

FORM PTO-1390 (Modified) (REV 11-2000)		U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE		ATTORNEY'S DOCKET NUMBER 209357US0XPCT	
TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371				U.S. APPLICATION NO. (IF KNOWN, SEE 37 CFR 09/869331	
INTERNATIONAL APPLICATION NO. PCT/JP00/00229		INTERNATIONAL FILING DATE 19 JANUARY 2000		PRIORITY DATE CLAIMED 29 JANUARY 1999 (earliest)	
TITLE OF INVENTION CATALYST FOR OLEFIN POLYMERIZATION AND PROCESS FOR PRODUCING OLEFIN POLYMER					
APPLICANT(S) FOR DO/EO/US Nobuhiro YABUNOUCHI, et al.					
Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:					
<ol style="list-style-type: none"> 1. <input checked="" type="checkbox"/> This is a FIRST submission of items concerning a filing under 35 U.S.C. 371. 2. <input type="checkbox"/> This is a SECOND or SUBSEQUENT submission of items concerning a filing under 35 U.S.C. 371. 3. <input checked="" type="checkbox"/> This is an express request to begin national examination procedures (35 U.S.C. 371(f)). The submission must include items (5), (6), (9) and (24) indicated below. 4. <input checked="" type="checkbox"/> The US has been elected by the expiration of 19 months from the priority date (Article 31). 5. <input checked="" type="checkbox"/> A copy of the International Application as filed (35 U.S.C. 371 (c) (2)) <ol style="list-style-type: none"> a. <input type="checkbox"/> is attached hereto (required only if not communicated by the International Bureau). b. <input checked="" type="checkbox"/> has been communicated by the International Bureau. c. <input type="checkbox"/> is not required, as the application was filed in the United States Receiving Office (RO/US). 6. <input checked="" type="checkbox"/> An English language translation of the International Application as filed (35 U.S.C. 371(c)(2)). <ol style="list-style-type: none"> a. <input checked="" type="checkbox"/> is attached hereto. b. <input type="checkbox"/> has been previously submitted under 35 U.S.C. 154(d)(4). 7. <input checked="" type="checkbox"/> Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371 (c)(3)) <ol style="list-style-type: none"> a. <input type="checkbox"/> are attached hereto (required only if not communicated by the International Bureau). b. <input type="checkbox"/> have been communicated by the International Bureau. c. <input type="checkbox"/> have not been made; however, the time limit for making such amendments has NOT expired. d. <input checked="" type="checkbox"/> have not been made and will not be made. 8. <input type="checkbox"/> An English language translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)). 9. <input checked="" type="checkbox"/> An oath or declaration of the inventor(s) (35 U.S.C. 371 (c)(4)). 10. <input type="checkbox"/> An English language translation of the annexes of the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371 (c)(5)). 11. <input checked="" type="checkbox"/> A copy of the International Preliminary Examination Report (PCT/IPEA/409). 12. <input checked="" type="checkbox"/> A copy of the International Search Report (PCT/ISA/210). <p>Items 13 to 20 below concern document(s) or information included:</p> <ol style="list-style-type: none"> 13. <input type="checkbox"/> An Information Disclosure Statement under 37 CFR 1.97 and 1.98. 14. <input type="checkbox"/> An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included. 15. <input checked="" type="checkbox"/> A FIRST preliminary amendment. 16. <input type="checkbox"/> A SECOND or SUBSEQUENT preliminary amendment. 17. <input type="checkbox"/> A substitute specification. 18. <input type="checkbox"/> A change of power of attorney and/or address letter. 19. <input type="checkbox"/> A computer-readable form of the sequence listing in accordance with PCT Rule 13ter.2 and 35 U.S.C. 1.821 - 1.825. 20. <input type="checkbox"/> A second copy of the published international application under 35 U.S.C. 154(d)(4). 21. <input type="checkbox"/> A second copy of the English language translation of the international application under 35 U.S.C. 154(d)(4). 22. <input type="checkbox"/> Certificate of Mailing by Express Mail 23. <input checked="" type="checkbox"/> Other items or information: <p>Request for Consideration of Documents in International Search Report Notice of Priority / PCT/IB/304 / PCT/IB/508</p>					

U.S. APPLICATION NO. (IF KNOWN, SEE 37 CFR 09/869331)		INTERNATIONAL APPLICATION NO. PCT/JP00/00229		ATTORNEY'S DOCKET NUMBER 209357US0XPCT	
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24. The following fees are submitted:

BASIC NATIONAL FEE (37 CFR 1.492 (a) (1) - (5)) :			CALCULATIONS PTO USE ONLY	
<input type="checkbox"/> Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO and International Search Report not prepared by the EPO or JPO	\$1000.00			
<input checked="" type="checkbox"/> International preliminary examination fee (37 CFR 1.482) not paid to USPTO but International Search Report prepared by the EPO or JPO	\$860.00			
<input type="checkbox"/> International preliminary examination fee (37 CFR 1.482) not paid to USPTO but international search fee (37 CFR 1.445(a)(2)) paid to USPTO	\$710.00			
<input type="checkbox"/> International preliminary examination fee (37 CFR 1.482) paid to USPTO but all claims did not satisfy provisions of PCT Article 33(1)-(4)	\$690.00			
<input type="checkbox"/> International preliminary examination fee (37 CFR 1.482) paid to USPTO and all claims satisfied provisions of PCT Article 33(1)-(4)	\$100.00			
ENTER APPROPRIATE BASIC FEE AMOUNT =		\$860.00		
Surcharge of \$130.00 for furnishing the oath or declaration later than months from the earliest claimed priority date (37 CFR 1.492 (e)). <input type="checkbox"/> 20 <input type="checkbox"/> 30		\$0.00		

CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE	
Total claims	40 - 20 =	20	x \$18.00	\$360.00
Independent claims	4 - 3 =	1	x \$80.00	\$80.00
Multiple Dependent Claims (check if applicable). <input type="checkbox"/>				\$0.00
TOTAL OF ABOVE CALCULATIONS =				\$1,300.00
<input type="checkbox"/> Applicant claims small entity status. (See 37 CFR 1.27). The fees indicated above are reduced by 1/2.				\$0.00
SUBTOTAL =				\$1,300.00
Processing fee of \$130.00 for furnishing the English translation later than months from the earliest claimed priority date (37 CFR 1.492 (f)). <input type="checkbox"/> 20 <input type="checkbox"/> 30 +				\$0.00
TOTAL NATIONAL FEE =				\$1,300.00
Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31) (check if applicable). <input type="checkbox"/>				\$0.00
TOTAL FEES ENCLOSED =				\$1,300.00
				Amount to be: refunded \$
				charged \$

a. ☒ A check in the amount of \$1,300.00 to cover the above fees is enclosed.


b. ☐ Please charge my Deposit Account No. _____ in the amount of _____ to cover the above fees. A duplicate copy of this sheet is enclosed.

c. ☒ The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. 15-0030. A duplicate copy of this sheet is enclosed.

d. ☐ Fees are to be charged to a credit card. **WARNING:** Information on this form may become public. **Credit card information should not be included on this form.** Provide credit card information and authorization on PTO-2038.

NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b)) must be filed and granted to restore the application to pending status.

SEND ALL CORRESPONDENCE TO:



22850

Surinder Sachar
Registration No. 34,423

(703) 413-3000

SIGNATURE

Norman F. Oblon
NAME

24,618
REGISTRATION NUMBER

7-26-01
DATE

209357US0XPCT

IN THE UNITED STATES PATENT & TRADEMARK OFFICE

IN RE APPLICATION OF: :
NOBUHIRO YABUNOUCHI ET AL : ATTN: APPLICATION DIVISION
SERIAL NO: U.S. PCT NEW APPLN :
(Based on PCT/JP00/00229)
FILED: HEREWITH : EXAMINER:
FOR: CATALYST FOR OLEFIN :
POLYMERIZATION AND
PROCESS FOR PRODUCING
OLEFIN POLYMER

PRELIMINARY AMENDMENT

ASSISTANT COMMISSIONER FOR PATENTS
WASHINGTON, D.C. 20231

SIR:

Prior to examination on the merits, please amend the above-identified application as follows:

IN THE CLAIMS

Please amend the claims as follows:

5. (Amended) The catalyst for polymerization of olefins as claimed in Claim 1, wherein at least one of three R¹'s is an aromatic hydrocarbon group having from 6 to 30 carbon atoms.

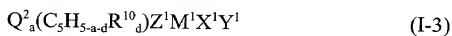
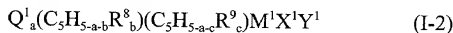
6. (Amended) The catalyst for polymerization of olefins as claimed in Claim 1, wherein three R¹'s are all aromatic hydrocarbon groups each having from 6 to 30 carbon atoms.

7. (Amended) The catalyst for polymerization of olefins as claimed in Claim 1, wherein three R¹'s are all phenyl groups.

8. (Amended) The catalyst for polymerization of olefins as claimed in Claim 1, wherein R² is an alkyl group having at least 2 carbon atoms.

9. (Amended) The catalyst for polymerization of olefins as claimed in Claim 1, wherein Z is aluminium.

10. (Amended) The catalyst for polymerization of olefins as claimed in Claim 1, wherein the transition metal compound (A) is represented by any of the following general formulae (I-2) to (I-6):



in which Q¹ represents a bonding group that crosslinks the two conjugated five-membered cyclic ligands (C₅H_{5-a-b}R⁸_b) and (C₅H_{5-a-c}R⁹_c); Q² represents a bonding group that crosslinks the conjugated five-membered cyclic ligand (C₅H_{5-a-d}R¹⁰_d) and the group Z¹; R⁸, R⁹, R¹⁰ and R¹¹ each represent a hydrocarbon group, a halogen atom, an alkoxy group, a silicon-containing hydrocarbon group, a phosphorus-containing hydrocarbon group, a nitrogen-containing hydrocarbon group, or a boron-containing hydrocarbon group; and a plurality of these groups, if any, may be the same or different, and may be bonded to each other to form a cyclic structure; a represents 0, 1 or 2; b, c and d each represent an integer of from 0 to 5 when a = 0, or an integer of from 0 to 4 when a = 1, or an integer of from 0 to 3 when a = 2; e is an integer of from 0 to 5; M¹ represents a transition metal of Groups 4 to 6 of the Periodic

Table; M² represents a transition metal of Groups 8 to 10 of the Periodic Table; L¹ and L² each represent a coordination-bonding ligand; X¹, Y¹, Z¹, W¹ and U¹ each represent a covalent-bonding or ionic-bonding ligand; and L¹, L², X¹, Y¹, Z¹, W¹ and U¹ may be bonded to each other to form a cyclic structure.

12. (Amended) A method for producing olefinic polymers, which comprises polymerizing olefins in the presence of the polymerization catalyst of Claim 1.

17. (Amended) The catalyst for polymerization of olefins as claimed in Claim 13, wherein at least one of three R³¹'s is an aromatic hydrocarbon group having from 6 to 30 carbon atoms.

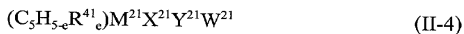
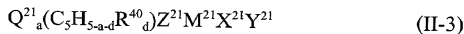
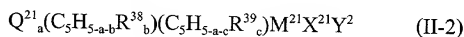
18. (Amended) The catalyst for polymerization of olefins as claimed in Claim 13, wherein three R³¹'s are all aromatic hydrocarbon groups each having from 6 to 30 carbon atoms.

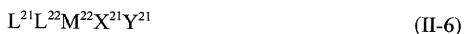
19. (Amended) The catalyst for polymerization of olefins as claimed in Claim 13, wherein three R³¹'s are all phenyl groups.

20. (Amended) The catalyst for polymerization of olefins as claimed in Claim 13, wherein R³² is an alkyl group having at least 2 carbon atoms.

21. (Amended) The catalyst for polymerization of olefins as claimed in Claim 15, wherein Z is aluminium.

22. (Amended) The catalyst for polymerization of olefins as claimed in Claim 13, wherein the transition metal compound (A) is represented by any of the following general formulae (II-2) to (II-6):





in which Q^{21} represents a bonding group that crosslinks the two conjugated five-membered cyclic ligands ($C_5H_{5-a-b}R^{38}_b$) and ($C_5H_{5-a-c}R^{39}_c$); Q^{22} represents a bonding group that crosslinks the conjugated five-membered cyclic ligand ($C_5H_{5-a-d}R^{40}_d$) and the group Z^{21} ; R^{38} , R^{39} , R^{40} and R^{41} each represent a hydrocarbon group, a halogen atom, an alkoxy group, a silicon-containing hydrocarbon group, a phosphorus-containing hydrocarbon group, a nitrogen-containing hydrocarbon group, or a boron-containing hydrocarbon group; and a plurality of these groups, if any, may be the same or different, and may be bonded to each other to form a cyclic structure; a represents 0, 1 or 2; b, c and d each represent an integer of from 0 to 5 when a = 0, or an integer of from 0 to 4 when a = 1, or an integer of from 0 to 3 when a = 2; e is an integer of from 0 to 5; M^{21} represents a transition metal of Groups 4 to 6 of the Periodic Table; M^{22} represents a transition metal of Groups 8 to 10 of the Periodic Table; L^{21} and L^{22} each represent a coordination-bonding ligand; X^{21} , Y^{21} , Z^{21} , W^{21} and U^{21} each represent a covalent-bonding or ionic-bonding ligand; and L^{21} , L^{22} , X^{21} , Y^{21} , Z^{21} , W^{21} and U^{21} may be bonded to each other to form a cyclic structure.

24. (Amended) A method for producing olefinic polymers, which comprises polymerizing olefins in the presence of the polymerization catalyst of Claim 13.

Please add new Claims 25-40 as follows:

25. (New) The catalyst for polymerization of olefins as claimed in Claim 4, wherein at least one of three R^1 's is an aromatic hydrocarbon group having from 6 to 30 carbon atoms.

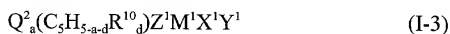
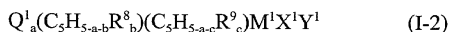
26. (New) The catalyst for polymerization of olefins as claimed in Claim 4, wherein three R^1 's are all aromatic hydrocarbon groups each having from 6 to 30 carbon atoms.

27. (New) The catalyst for polymerization of olefins as claimed in Claim 4, wherein three R¹'s are all phenyl groups.

28. (New) The catalyst for polymerization of olefins as claimed in Claim 4, wherein R² is an alkyl group having at least 2 carbon atoms.

29. (New) The catalyst for polymerization of olefins as claimed in Claim 4, wherein Z is aluminium.

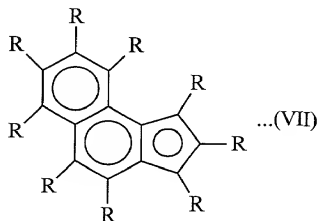
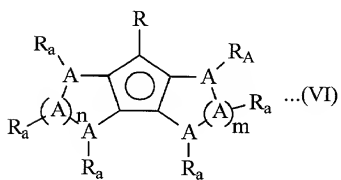
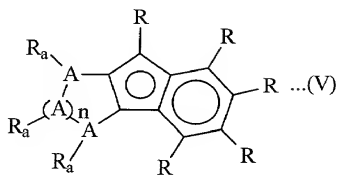
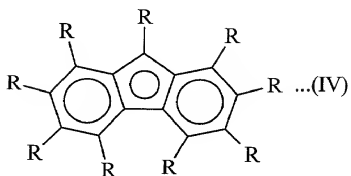
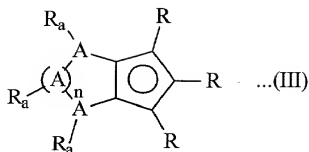
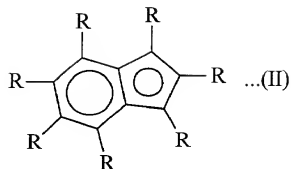
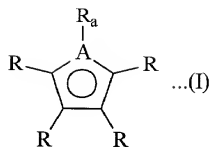
30. (New) The catalyst for polymerization of olefins as claimed in Claim 4, wherein the transition metal compound (A) is represented by any of the following general formulae (I-2) to (I-6):



in which Q¹ represents a bonding group that crosslinks the two conjugated five-membered cyclic ligands (C₅H_{5-a-b}R⁸_b) and (C₅H_{5-a-c}R⁹_c); Q² represents a bonding group that crosslinks the conjugated five-membered cyclic ligand (C₅H_{5-a-d}R¹⁰_d) and the group Z¹; R⁸, R⁹, R¹⁰ and R¹¹ each represent a hydrocarbon group, a halogen atom, an alkoxy group, a silicon-containing hydrocarbon group, a phosphorus-containing hydrocarbon group, a nitrogen-containing hydrocarbon group, or a boron-containing hydrocarbon group; and a plurality of these groups, if any, may be the same or different, and may be bonded to each other to form a cyclic structure; a represents 0, 1 or 2; b, c and d each represent an integer of from 0 to 5 when a = 0, or an integer of from 0 to 4 when a = 1, or an integer of from 0 to 3 when a = 2; e is an integer of from 0 to 5; M¹ represents a transition metal of Groups 4 to 6 of the Periodic

Table; M^2 represents a transition metal of Groups 8 to 10 of the Periodic Table; L^1 and L^2 each represent a coordination-bonding ligand; X^1 , Y^1 , Z^1 , W^1 and U^1 each represent a covalent-bonding or ionic-bonding ligand; and L^1 , L^2 , X^1 , Y^1 , Z^1 , W^1 and U^1 may be bonded to each other to form a cyclic structure.

31. (New) The catalyst for polymerization of olefins as claimed in Claim 30, wherein, in the transition metal compound (A) of formula (I-4), the group $(C_5H_{5-\sigma}R^{II})_\sigma$ is represented by any of the following general formulae (I) to (VII):



wherein A represents an element of Group 13, 14, 15 or 16, and plural A's may be the same or different; R represents a hydrogen atom, a halogen atom, an aliphatic hydrocarbon group having from 1 to 30 carbon atoms, an aromatic hydrocarbon group having from 6 to 30

carbon atoms, an alkoxy group having from 1 to 30 carbon atoms, an aryloxy group having from 6 to 30 carbon atoms, a thioalkoxy group having from 1 to 30 carbon atoms, a thioaryloxy group having from 6 to 30 carbon atoms, an amino group, an amido group, a carboxyl group, or an alkylsilyl or alkylsilylalkyl group having from 3 to 30 carbon atoms, and R's may be the same or different, and may be optionally bonded to each other to form a cyclic structure; a represents 0, 1 or 2; and n and m each represent an integer of at least 1.

32. (New) A method for producing olefinic polymers, which comprises polymerizing olefins in the presence of the polymerization catalyst of Claim 4.

33. (New) The catalyst for polymerization of olefins as claimed in Claim 16, wherein at least one of three R³¹'s is an aromatic hydrocarbon group having from 6 to 30 carbon atoms.

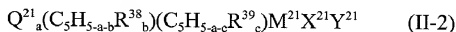
34. (New) The catalyst for polymerization of olefins as claimed in Claim 16, wherein three R³¹'s are all aromatic hydrocarbon groups each having from 6 to 30 carbon atoms.

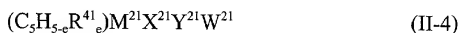
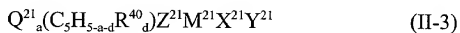
35. (New) The catalyst for polymerization of olefins as claimed in Claim 16, wherein three R³¹'s are all phenyl groups.

36. (New) The catalyst for polymerization of olefins as claimed in Claim 16, wherein R³² is an alkyl group having at least 2 carbon atoms.

37. (New) The catalyst for polymerization of olefins as claimed in Claim 16, wherein Z is aluminium.

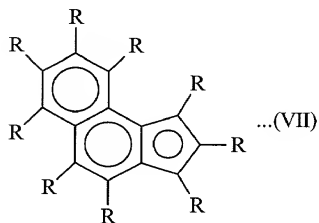
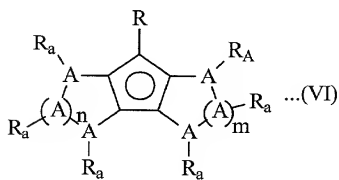
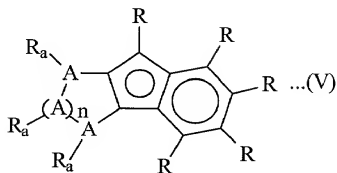
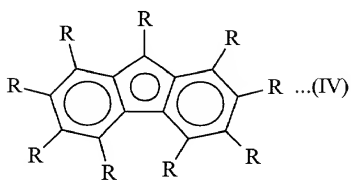
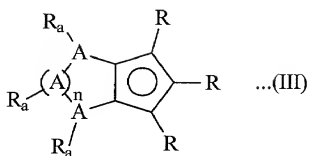
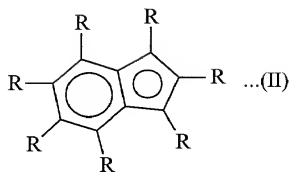
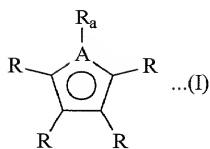
38. (New) The catalyst for polymerization of olefins as claimed in Claim 16, wherein the transition metal compound (A) is represented by any of the following general formulae (II-2) to (II-6):





in which Q^{21} represents a bonding group that crosslinks the two conjugated five-membered cyclic ligands ($C_5H_{5-a-b}R^{38}_b$) and ($C_5H_{5-a-c}R^{39}_c$); Q^{22} represents a bonding group that crosslinks the conjugated five-membered cyclic ligand ($C_5H_{5-a-d}R^{40}_d$) and the group Z^{21} ; R^{38} , R^{39} , R^{40} and R^{41} each represent a hydrocarbon group, a halogen atom, an alkoxy group, a silicon-containing hydrocarbon group, a phosphorus-containing hydrocarbon group, a nitrogen-containing hydrocarbon group, or a boron-containing hydrocarbon group; and a plurality of these groups, if any, may be the same or different, and may be bonded to each other to form a cyclic structure; a represents 0, 1 or 2; b, c and d each represent an integer of from 0 to 5 when a = 0, or an integer of from 0 to 4 when a = 1, or an integer of from 0 to 3 when a = 2; e is an integer of from 0 to 5; M^{21} represents a transition metal of Groups 4 to 6 of the Periodic Table; M^{22} represents a transition metal of Groups 8 to 10 of the Periodic Table; L^{21} and L^{22} each represent a coordination-bonding ligand; X^{21} , Y^{21} , Z^{21} , W^{21} and U^{21} each represent a covalent-bonding or ionic-bonding ligand; and L^{21} , L^{22} , X^{21} , Y^{21} , Z^{21} , W^{21} and U^{21} may be bonded to each other to form a cyclic structure.

39. (New) The catalyst for polymerization of olefins as claimed in Claim 38, wherein, in the transition metal compound (A) of formula (II-4), the group ($C_5H_{5-e}R^{41}_e$) is represented by any of the following general formulae (I) to (VII):



wherein A represents an element of Group 13, 14, 15 or 16, and plural A's may be the same or different; R represents a hydrogen atom, a halogen atom, an aliphatic hydrocarbon group having from 1 to 30 carbon atoms, an aromatic hydrocarbon group having from 6 to 30 carbon atoms, an alkoxy group having from 1 to 30 carbon atoms, an aryloxy group having from 6 to 30 carbon atoms, a thioalkoxy group having from 1 to 30 carbon atoms, a

thioaryloxy group having from 6 to 30 carbon atoms, an amino group, an amido group, a carboxyl group, or an alkylsilyl or alkylsilylalkyl group having from 3 to 30 carbon atoms, and R's may be the same or different, and may be optionally bonded to each other to form a cyclic structure; a represents 0, 1 or 2; and n and m each represent an integer of at least 1.


40. (New) A method for producing olefinic polymers, which comprises polymerizing olefins in the presence of the polymerization catalyst of Claim 16.

REMARKS

Claims 1-40 are active in the present application. Claims 5-10, 12, 17-22, and 24 have been amended to remove multiple dependencies. New Claims 25-40 have been added. Support for new Claims 25-40 is found in the original Claims 1-24. No new matter is added. An action on the merits and allowance of claims is solicited.

Respectfully submitted,

OBLON, SPIVAK, McCLELLAND,
MAIER & NEUSTADT, P.C.


Norman F. Oblon
Attorney of Record
Registration No. 24,618

Daniel J. Pereira
Registration No. 45,518



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(703) 413-3000
Fax #: (703) 413-2220
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Serial No: _____

Amendment Filed on: _____

07-26-01

5. (Amended) The catalyst for polymerization of olefins as claimed in [any of claims 1 to 4] Claim 1, wherein at least one of three R¹'s is an aromatic hydrocarbon group having from 6 to 30 carbon atoms.

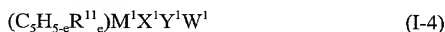
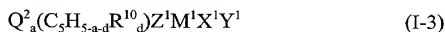
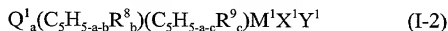
6. (Amended) The catalyst for polymerization of olefins as claimed in [any of claims 1 to 4] Claim 1, wherein three R¹'s are all aromatic hydrocarbon groups each having from 6 to 30 carbon atoms.

7. (Amended) The catalyst for polymerization of olefins as claimed in [any of claims 1 to 4] Claim 1, wherein three R¹'s are all phenyl groups.

8. (Amended) The catalyst for polymerization of olefins as claimed in [any of claims 1 to 7] Claim 1, wherein R² is an alkyl group having at least 2 carbon atoms.

9. (Amended) The catalyst for polymerization of olefins as claimed in [any of claims 3 to 8] Claim 1, wherein Z is aluminium.

10. (Amended) The catalyst for polymerization of olefins as claimed in [any of claims 1 to 9] Claim 1, wherein the transition metal compound (A) is represented by any of the following general formulae (I-2) to (I-6):



in which Q¹ represents a bonding group that crosslinks the two conjugated five-membered cyclic ligands (C₅H_{5-a-b}R⁸_b) and (C₅H_{5-a-c}R⁹_c); Q² represents a bonding group that crosslinks the conjugated five-membered cyclic ligand (C₅H_{5-a-d}R¹⁰_d) and the group Z¹; R⁸, R⁹, R¹⁰ and R¹¹ each represent a hydrocarbon group, a halogen atom, an alkoxy group, a silicon-containing hydrocarbon group, a phosphorus-containing hydrocarbon group, a nitrogen-containing hydrocarbon group, or a boron-containing hydrocarbon group; and a plurality of these groups, if any, may be the same or different, and may be bonded to each other to form a cyclic structure; a represents 0, 1 or 2; b, c and d each represent an integer of from 0 to 5 when a = 0, or an integer of from 0 to 4 when a = 1, or an integer of from 0 to 3 when a = 2; e is an integer of from 0 to 5; M¹ represents a transition metal of Groups 4 to 6 of the Periodic Table; M² represents a transition metal of Groups 8 to 10 of the Periodic Table; L¹ and L² each represent a coordination-bonding ligand; X¹, Y¹, Z¹, W¹ and U¹ each represent a covalent-bonding or ionic-bonding ligand; and L¹, L², X¹, Y¹, Z¹, W¹ and U¹ may be bonded to each other to form a cyclic structure.

12. (Amended) A method for producing olefinic polymers, which comprises polymerizing olefins in the presence of the polymerization catalyst of [any of claims 1 to 11] Claim 1.

17. (Amended) The catalyst for polymerization of olefins as claimed in [any of claims 13 to 16] Claim 13, wherein at least one of three R³¹'s is an aromatic hydrocarbon group having from 6 to 30 carbon atoms.

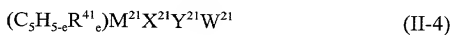
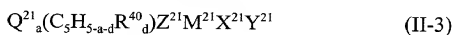
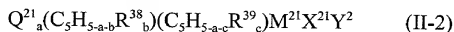
18. (Amended) The catalyst for polymerization of olefins as claimed in [any of claims 13 to 16] Claim 13, wherein three R³¹'s are all aromatic hydrocarbon groups each having from 6 to 30 carbon atoms.

19. (Amended) The catalyst for polymerization of olefins as claimed in [any of claims 13 to 16] Claim 13, wherein three R³¹'s are all phenyl groups.

20. (Amended) The catalyst for polymerization of olefins as claimed in [any of claims 13 to 19] Claim 13, wherein R³² is an alkyl group having at least 2 carbon atoms.

21. (Amended) The catalyst for polymerization of olefins as claimed in [any of claims 15 to 20] Claim 15, wherein Z is aluminium.

22. (Amended) The catalyst for polymerization of olefins as claimed in [any of claims 13 to 21] Claim 13, wherein the transition metal compound (A) is represented by any of the following general formulae (II-2) to (II-6):



in which Q²¹ represents a bonding group that crosslinks the two conjugated five-membered cyclic ligands (C₅H_{5-a-b}R³⁸_b) and (C₅H_{5-a-c}R³⁹_c); Q²² represents a bonding group that crosslinks the conjugated five-membered cyclic ligand (C₅H_{5-a-d}R⁴⁰_d) and the group Z²¹; R³⁸, R³⁹, R⁴⁰ and R⁴¹ each represent a hydrocarbon group, a halogen atom, an alkoxy group, a silicon-containing hydrocarbon group, a phosphorus-containing hydrocarbon group, a nitrogen-containing hydrocarbon group, or a boron-containing hydrocarbon group; and a plurality of these groups, if any, may be the same or different, and may be bonded to each other to form a cyclic structure; a represents 0, 1 or 2; b, c and d each represent an integer of from 0 to 5 when a = 0, or an integer of from 0 to 4 when a = 1, or an integer of from 0 to 3 when a = 2; e is an integer of from 0 to 5; M²¹ represents a transition metal of Groups 4 to 6

of the Periodic Table; M^{22} represents a transition metal of Groups 8 to 10 of the Periodic Table; L^{21} and L^{22} each represent a coordination-bonding ligand; X^{21} , Y^{21} , Z^{21} , W^{21} and U^{21} each represent a covalent-bonding or ionic-bonding ligand; and L^{21} , L^{22} , X^{21} , Y^{21} , Z^{21} , W^{21} and U^{21} may be bonded to each other to form a cyclic structure.

24. (Amended) A method for producing olefinic polymers, which comprises polymerizing olefins in the presence of the polymerization catalyst of [any of claims 13 to 23]

Claim 13.

Claims 25-40 (New).

DESCRIPTION

CATALYST FOR OLEFIN POLYMERIZATION AND PROCESS FOR PRODUCING
OLEFIN POLYMER

TECHNICAL FIELD

The present invention relates to catalysts for polymerization of olefins, and to methods for producing olefinic polymers. Precisely, it relates to catalysts for olefin polymerization comprising, as one component, a specific compound, and to inexpensive and efficient methods for producing olefinic polymers with the catalysts.

BACKGROUND ART

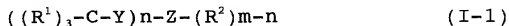
Recently, metallocene catalysts have been developed and used for olefinic polymer production, which comprise, as the catalyst component, a transition metal compound having a π -ligand bonded to the center metal element via a group.

To exhibit satisfactory activity, however, the catalysts of that type require a large amount of promoters such as aluminoxanes and the like. Therefore, they are problematic in that the total catalyst costs are inevitably high, and, in addition, the catalyst residue resulting from the promoter used often remains in the polymers produced thereby unfavorably coloring the polymers.

In that situation, techniques of using clay, clay

1. A catalyst for polymerization of olefins, which comprises:

- (A) a transition metal compound,
- (B) an oxygen-containing compound,
- (C) a compound of a general formula (I-1):



wherein R^1 represents an aromatic hydrocarbon group having from 6 to 30 carbon atoms, an alkoxy group having from 1 to 30 carbon atoms, an aryloxy group having from 6 to 30 carbon atoms, a thioalkoxy group having from 1 to 30 carbon atoms, a thioaryloxy group having from 6 to 30 carbon atoms, an amino group, an amido group, or a carboxyl group, R^1 's may be the same or different, and R^1 's may be optionally bonded to each other to form a cyclic structure; Y represents an element of Group 16; Z represents a metal element of Groups 2 to 13; R^2 represents a hydrocarbon group; m is an integer, indicating the valency of the metal element Z; and n is an integer of from 1 to (m-1),

and optionally,

- (D) an alkylating agent.

2. The catalyst of above 1 for polymerization of olefins, wherein, in (C), Y is oxygen and Z is aluminium.

3. The catalyst of above 1 for polymerization of olefins, wherein the compound (C) is a reaction product of a compound of a general formula, $(R^1)_3-C-OR^3$, and a compound of

a general formula, $Z(R^2)_m$:

in which R^1 represents an aromatic hydrocarbon group having from 6 to 30 carbon atoms, an alkoxy group having from 1 to 30 carbon atoms, an aryloxy group having from 6 to 30 carbon atoms, a thioalkoxy group having from 1 to 30 carbon atoms, a thioaryloxy group having from 6 to 30 carbon atoms, an amino group, an amido group, or a carboxyl group, R^1 's may be the same or different, and R^1 's may be optionally bonded to each other to form a cyclic structure; R^3 represents a hydrogen atom, a halogen atom, an aliphatic hydrocarbon group having from 1 to 30 carbon atoms, an aromatic hydrocarbon group having from 6 to 30 carbon atoms, an alkoxy group having from 1 to 30 carbon atoms, an aryloxy group having from 6 to 30 carbon atoms, a thioalkoxy group having from 1 to 30 carbon atoms, a thioaryloxy group having from 6 to 30 carbon atoms, an amino group, an amido group, or a carboxyl group; Z represents a metal element of Groups 2 to 13; m is an integer, indicating the valency of the metal element Z ; and R^2 represents a hydrocarbon group.

4. A catalyst for polymerization of olefins, which comprises:

(A) a transition metal compound,

(B) an oxygen-containing compound,

(C1) a compound of a general formula, $(R^1)_3-C-OR^3$:

wherein R^1 represents an aromatic hydrocarbon group having

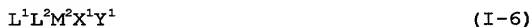
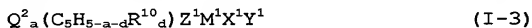
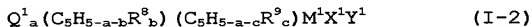
of olefins, wherein three R¹'s are all aromatic hydrocarbon groups each having from 6 to 30 carbon atoms.

7. The catalyst of any of above 1 to 4 for polymerization of olefins, wherein three R¹'s are all phenyl groups.

8. The catalyst of any of above 1 to 7 for polymerization of olefins, wherein R² is an alkyl group having at least 2 carbon atoms.

9. The catalyst of any of above 3 to 8 for polymerization of olefins, wherein Z is aluminium.

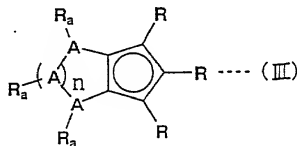
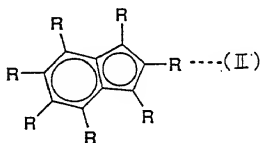
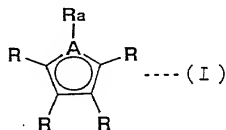
10. The catalyst of any of above 1 to 9 for polymerization of olefins, wherein the transition metal compound (A) is represented by any of the following general formulae (I-2) to (I-6):

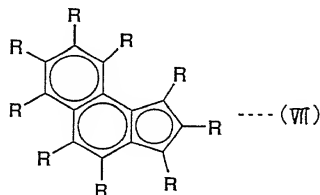
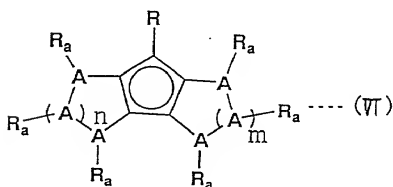
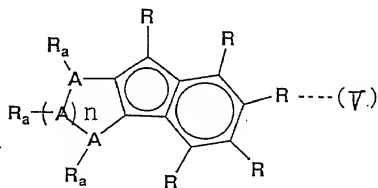
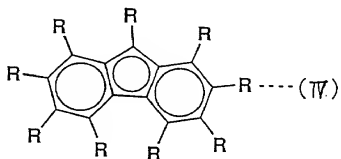


in which Q¹ represents a bonding group that crosslinks the two conjugated five-membered cyclic ligands (C₅H_{5-a-b}R⁸_b) and (C₅H_{5-a-c}R⁹_c); Q² represents a bonding group that crosslinks the conjugated five-membered cyclic ligand (C₅H_{5-a-d}R¹⁰_d) and the group Z¹; R⁸, R⁹, R¹⁰ and R¹¹ each represent a hydrocarbon group, a halogen atom, an alkoxy group, a silicon-containing hydrocarbon group, a phosphorus-containing

hydrocarbon group, a nitrogen-containing hydrocarbon group, or a boron-containing hydrocarbon group; and a plurality of these groups, if any, may be the same or different, and may be bonded to each other to form a cyclic structure; a represents 0, 1 or 2; b, c and d each represent an integer of from 0 to 5 when a = 0, or an integer of from 0 to 4 when a = 1, or an integer of from 0 to 3 when a = 2; e is an integer of from 0 to 5; M¹ represents a transition metal of Groups 4 to 6 of the Periodic Table; M² represents a transition metal of Groups 8 to 10 of the Periodic Table; L¹ and L² each represent a coordination-bonding ligand; X¹, Y¹, Z¹, W¹ and U¹ each represent a covalent-bonding or ionic-bonding ligand; and L¹, L², X¹, Y¹, Z¹, W¹ and U¹ may be bonded to each other to form a cyclic structure.

11. The catalyst of above 10 for polymerization of olefins, wherein, in the transition metal compound (A) of formula (I-4), the group (C₅H_{5-e}R^{11e}) is represented by any of the following general formulae (I) to (VII):





wherein A represents an element of Group 13, 14, 15 or 16, and plural A's may be the same or different; R represents a hydrogen atom, a halogen atom, an aliphatic hydrocarbon group having from 1 to 30 carbon atoms, an aromatic hydrocarbon group having from 6 to 30 carbon atoms, an alkoxy group having from 1 to 30 carbon atoms, an aryloxy group having from 6 to 30 carbon atoms, a thioalkoxy group having from 1 to 30 carbon atoms, a thioaryloxy group having from 6 to 30 carbon atoms, an amino group, an amido group, a carboxyl group, or an alkylsilyl or alkylsilylalkyl group having from 3 to 30 carbon atoms, and R's may be the same or different, and may be optionally bonded to each other to form a cyclic structure; a represents 0, 1 or 2; and n

and m each represent an integer of at least 1.

12. A method for producing olefinic polymers, which comprises polymerizing olefins in the presence of the polymerization catalyst of any of above 1 to 11.

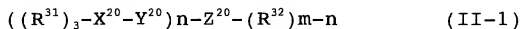
Another aspect of the invention is to provide a catalyst for polymerization of olefins and a method for producing olefinic polymers, which are as follows:

1. A catalyst for polymerization of olefins, which comprises:

(A) a transition metal compound,

(B) a compound capable of reacting with a transition metal compound to form an ionic complex,

(C) a compound of a general formula (II-1):



wherein R^{31} represents a hydrogen atom, a halogen atom, an aliphatic hydrocarbon group having from 1 to 30 carbon atoms, an aromatic hydrocarbon group having from 6 to 30 carbon atoms, an alkoxy group having from 1 to 30 carbon atoms, an aryloxy group having from 6 to 30 carbon atoms, a thioalkoxy group having from 1 to 30 carbon atoms, a thioaryloxy group having from 6 to 30 carbon atoms, an amino group, an amido group, or a carboxyl group, R^{31} 's may be the same or different, and R^{31} 's may be optionally bonded to each other to form a cyclic structure; X^{20} represents an element of Group 14; Y^{20} represents an element of Group

16; Z^{20} represents a metal element of Groups 2 to 13; R^{32} represents a hydrocarbon group; m is an integer, indicating the valency of the metal element Z^{20} ; and n is an integer of from 1 to $(m-1)$,

and optionally,

(D) an alkylating agent.

2. The catalyst of above 1 for polymerization of olefins, wherein, in (C), Y^{20} is oxygen and Z^{20} is aluminium.

3. The catalyst of above 1 for polymerization of olefins, wherein the compound (C) is a reaction product of a compound of a general formula, $(R^{31})_3-C-OR^{33}$, and a compound of a general formula, $Z^{20}(R^{32})_m$:

in which R^{31} represents a hydrogen atom, a halogen atom, an aliphatic hydrocarbon group having from 1 to 30 carbon atoms, an aromatic hydrocarbon group having from 6 to 30 carbon atoms, an alkoxy group having from 1 to 30 carbon atoms, an aryloxy group having from 6 to 30 carbon atoms, a thioalkoxy group having from 1 to 30 carbon atoms, a thioaryloxy group having from 6 to 30 carbon atoms, an amino group, an amido group, or a carboxyl group, R^{31} 's may be the same or different, and R^{31} 's may be optionally bonded to each other to form a cyclic structure; Z^{20} represents a metal element of Groups 2 to 13; R^{32} represents a hydrocarbon group; R^{33} represents a hydrogen atom, a halogen atom, an aliphatic hydrocarbon group having from 1 to 30

carbon atoms, an aromatic hydrocarbon group having from 6 to 30 carbon atoms, an alkoxy group having from 1 to 30 carbon atoms, an aryloxy group having from 6 to 30 carbon atoms, a thioalkoxy group having from 1 to 30 carbon atoms, a thioaryloxy group having from 6 to 30 carbon atoms, an amino group, an amido group, or a carboxyl group, and may be the same or different; m is an integer, indicating the valency of the metal element Z^{20} ; and n is an integer of from 1 to (m-1).

4. A catalyst for polymerization of olefins, which comprises:

(A) a transition metal compound,

(B) a compound capable of reacting with a transition metal compound to form an ionic complex,

(C1) a compound of a general formula, $(R^{31})_3-C-OR^{33}$:

wherein R^{31} represents a hydrogen atom, a halogen atom, an aliphatic hydrocarbon group having from 1 to 30 carbon atoms, an aromatic hydrocarbon group having from 6 to 30 carbon atoms, an alkoxy group having from 1 to 30 carbon atoms, an aryloxy group having from 6 to 30 carbon atoms, a thioalkoxy group having from 1 to 30 carbon atoms, a thioaryloxy group having from 6 to 30 carbon atoms, an amino group, an amido group, or a carboxyl group, R^{31} 's may be the same or different, and R^{31} 's may be optionally bonded to each other to form a cyclic structure; R^{33} represents

a hydrogen atom, a halogen atom, an aliphatic hydrocarbon group having from 1 to 30 carbon atoms, an aromatic hydrocarbon group having from 6 to 30 carbon atoms, an alkoxy group having from 1 to 30 carbon atoms, an aryloxy group having from 6 to 30 carbon atoms, a thioalkoxy group having from 1 to 30 carbon atoms, a thioaryloxy group having from 6 to 30 carbon atoms, an amino group, an amido group, or a carboxyl group, and may be the same or different,

(C2) a compound of a general formula, $Z^{20}(R^{32})_m$, wherein Z^{20} represents a metal element of Groups 2 to 13; m is an integer, indicating the valency of the metal element Z^{20} ; and R^{32} represents a hydrocarbon group,

and optionally,

(D) an alkylating agent.

5. The catalyst of any of above 1 to 4 for polymerization of olefins, wherein at least one of three R^{31} 's is an aromatic hydrocarbon group having from 6 to 30 carbon atoms.

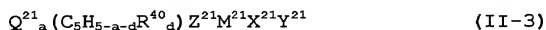
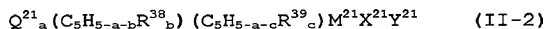
6. The catalyst of any of above 1 to 4 for polymerization of olefins, wherein three R^{31} 's are all aromatic hydrocarbon groups each having from 6 to 30 carbon atoms.

7. The catalyst of any of above 1 to 4 for polymerization of olefins, wherein three R^{31} 's are all phenyl groups.

8. The catalyst of any of above 1 to 7 for polymerization of olefins, wherein R^{32} is an alkyl group having at least 2 carbon atoms.

9. The catalyst of any of above 3 to 8 for polymerization of olefins, wherein Z is aluminium.

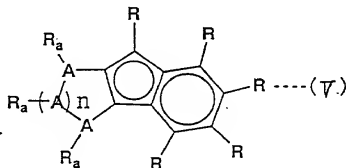
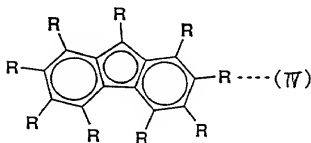
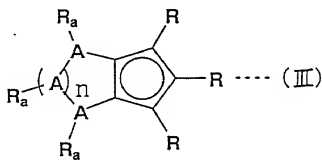
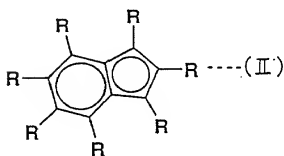
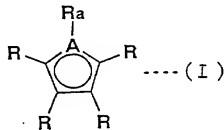
10. The catalyst of any of above 1 to 9 for polymerization of olefins, wherein the transition metal compound (A) is represented by any of the following general formulae (II-2) to (II-6):

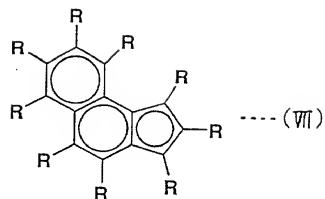
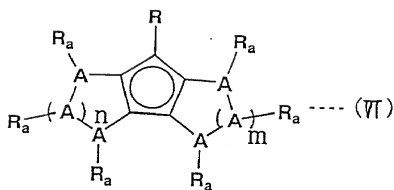


in which Q^{21} represents a bonding group that crosslinks the two conjugated five-membered cyclic ligands $(C_5H_{5-a-b}R^{38}_b)$ and $(C_5H_{5-a-c}R^{39}_c)$; Q^{22} represents a bonding group that crosslinks the conjugated five-membered cyclic ligand $(C_5H_{5-a-d}R^{40}_d)$ and the group Z^{21} ; R^{38} , R^{39} , R^{40} and R^{41} each represent a hydrocarbon group, a halogen atom, an alkoxy group, a silicon-containing hydrocarbon group, a phosphorus-containing hydrocarbon group, a nitrogen-containing hydrocarbon group, or a boron-containing hydrocarbon group; and a plurality of these groups, if any, may be the same or different, and may be bonded to each other to form a cyclic structure; a represents 0, 1 or 2; b, c and d each represent an integer of from 0 to 5 when a = 0, or an integer of from 0 to 4 when a = 1, or an integer of

from 0 to 3 when $a = 2$; e is an integer of from 0 to 5; M^{21} represents a transition metal of Groups 4 to 6 of the Periodic Table; M^{22} represents a transition metal of Groups 8 to 10 of the Periodic Table; L^{21} and L^{22} each represent a coordination-bonding ligand; X^{21} , Y^{21} , Z^{21} , W^{21} and U^{21} each represent a covalent-bonding or ionic-bonding ligand; and L^{21} , L^{22} , X^{21} , Y^{21} , Z^{21} , W^{21} and U^{21} may be bonded to each other to form a cyclic structure.

11. The catalyst of above 10 for polymerization of olefins, wherein, in the transition metal compound (A) of formula (II-4), the group $(C_5H_{5-e}R^{41}_e)$ is represented by any of the following general formulae (I) to (VII):





wherein A represents an element of Group 13, 14, 15 or 16, and plural A's may be the same or different; R represents a hydrogen atom, a halogen atom, an aliphatic hydrocarbon group having from 1 to 30 carbon atoms, an aromatic hydrocarbon group having from 6 to 30 carbon atoms, an alkoxy group having from 1 to 30 carbon atoms, an aryloxy group having from 6 to 30 carbon atoms, a thioalkoxy group having from 1 to 30 carbon atoms, a thioaryloxy group having from 6 to 30 carbon atoms, an amino group, an amido group, a carboxyl group, or an alkylsilyl or alkylsilylalkyl group having from 3 to 30 carbon atoms, and R's may be the same or different, and may be optionally bonded to each other to form a cyclic structure; a represents 0, 1 or 2; and n and m each represent an integer of at least 1.

12. A method for producing olefinic polymers, which comprises polymerizing olefins in the presence of the polymerization catalyst of any of above 1 to 11.

BEST MODES OF CARRYING OUT THE INVENTION

The first and second aspects of the invention are

described below with reference to their embodiments.

[First aspect of the invention]

In this section, the first aspect of the invention will be simply referred to as "the invention".

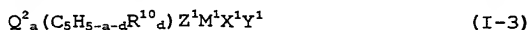
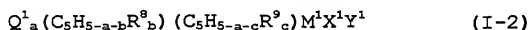
I. Catalyst for polymerization of olefins:

1. Components of catalyst for polymerization of olefins:

The catalyst of the invention for polymerization of olefins comprises (A) a transition metal compound, (B) an oxygen-containing compound, and (C) a compound of formula (I) which will be described in detail hereinunder, and optionally (D) an alkylating agent. The constituent components are described below.

(1) (A) Transition metal compound:

Various types of transition metal compounds are usable as the component (A) in the invention, but preferred are transition metal compounds of Groups 4 to 6 of the Periodic Table, or transition metal compounds of Groups 8 to 10 thereof. As the transition metal compounds of Groups 4 to 6 of the Periodic Table, preferred are those of the following general formulae (I-2) to (I-5). As the transition metal compounds of Groups 8 to 10 of the Periodic Table, preferred are those of the following general formula (I-6).



M¹ represents a transition metal element of Groups 4 to 6 of the Periodic Table, including, for example, titanium, zirconium, hafnium, niobium, molybdenum, tungsten, etc. Of those, preferred are titanium, zirconium and hafnium, and more preferred is zirconium. Z¹ represents a covalent-bonding ligand, including, for example, oxygen (-O-), sulfur (-S-), an alkoxy group having from 1 to 20, preferably from 1 to 10 carbon atoms, a thioalkoxy group having from 1 to 20, preferably from 1 to 12 carbon atoms, a nitrogen-containing hydrocarbon group having from 1 to 40, preferably from 1 to 18 carbon atoms, and a phosphorus-containing hydrocarbon group having from 1 to 40, preferably from 1 to 18 carbon atoms. X¹ and Y¹ each represent a covalent-bonding ligand, including, for example, a hydrogen atom, a halogen atom, a hydrocarbon group having from 1 to 20, preferably from 1 to 10 carbon atoms, an alkoxy group having from 1 to 20, preferably from 1 to 10 carbon atoms, an amino group, a phosphorus-containing hydrocarbon group having from 1 to 20, preferably from 1 to 12 carbon atoms (e.g., a diphenylphosphine group, etc.), a silicon-containing hydrocarbon group having from 1 to 20, preferably from 1 to 12 carbon atoms (e.g., a trimethylsilyl group, etc.), and a boron compound residue having a hydrocarbon group with from 1 to 20, preferably from 1 to 12 carbon atoms or having halogens (e.g., B(C₆H₅)₄, BF₄). Of those, preferred are halogen atoms

and hydrocarbon groups. X^1 and Y^1 may be the same or different, and they may be bonded to each other to form a cyclic structure.

In formula (I-4), M^1 represents a transition metal of Groups 4 to 6 of the Periodic Table, such as that mentioned above; and X^1 and Y^1 have the same meanings as above. The meanings of X^1 and Y^1 shall apply to W^1 . Namely, W^1 is a covalent-bonding ligand, including, for example, a hydrogen atom, a halogen atom, a hydrocarbon group having from 1 to 20, preferably from 1 to 10 carbon atoms, an alkoxy group having from 1 to 20, preferably from 1 to 10 carbon atoms, an amino group, a phosphorus-containing hydrocarbon group having from 1 to 20, preferably from 1 to 12 carbon atoms (e.g., a diphenylphosphine group, etc.), a silicon-containing hydrocarbon group having from 1 to 20, preferably from 1 to 12 carbon atoms (e.g., a trimethylsilyl group, etc.), and a boron compound residue having a hydrocarbon group with from 1 to 20, preferably from 1 to 12 carbon atoms or having halogens (e.g., $B(C_6H_5)_4$, BF_4). Of those, preferred are halogen atoms and hydrocarbon groups. X^1 , Y^1 and W^1 may be the same or different, and they may be bonded to each other to form a cyclic structure.

In formula (I-5), M^1 represents a transition metal of Groups 4 to 6 of the Periodic Table, such as that mentioned above; and X^1 , Y^1 and W^1 have the same meanings as above. The meanings of X^1 , Y^1 and W^1 shall apply to U^1 . Namely, U^1 is a

covalent-bonding ligand, including, for example, a hydrogen atom, a halogen atom, a hydrocarbon group having from 1 to 20, preferably from 1 to 10 carbon atoms, an alkoxy group having from 1 to 20, preferably from 1 to 10 carbon atoms, an amino group, a phosphorus-containing hydrocarbon group having from 1 to 20, preferably from 1 to 12 carbon atoms (e.g., a diphenylphosphine group, etc.), a silicon-containing hydrocarbon group having from 1 to 20, preferably from 1 to 12 carbon atoms (e.g., a trimethylsilyl group, etc.), and a boron compound residue having a hydrocarbon group with from 1 to 20, preferably from 1 to 12 carbon atoms or having halogens (e.g., $B(C_6H_5)_4$, BF_4). Of those, preferred are halogen atoms and hydrocarbon groups. X^1 , Y^1 , W^1 and U^1 may be the same or different, and they may be bonded to each other to form a cyclic structure.

(I) As specific examples of the transition metal compounds of formulae (I-2) and (I-3), mentioned are the following compounds. In the compounds to be mentioned below, titanium may be replaced with zirconium, and such zirconium compounds are also referred to herein to the same effect.

(1) Transition metal compounds not having a crosslinkable bonding group but having two conjugated, 5-membered cyclic ligands, such as
 bis(cyclopentadienyl)titanium dichloride,
 bis(methylcyclopentadienyl)titanium dichloride,

bis(dimethylcyclopentadienyl)titanium	dichloride,
bis(trimethylcyclopentadienyl)titanium	dichloride,
bis(tetramethylcyclopentadienyl)titanium	dichloride,
bis(pentamethylcyclopentadienyl)titanium	dichloride,
bis(n-butylcyclopentadienyl)titanium	dichloride,
bis(indenyl)titanium	dichloride, bis(fluorenyl)titanium
dichloride, bis(cyclopentadienyl)titanium	chlorohydride,
bis(cyclopentadienyl)methyltitanium	chloride,
bis(cyclopentadienyl)ethyltitanium	chloride,
bis(cyclopentadienyl)phenyltitanium	chloride,
bis(cyclopentadienyl)dimethyltitanium,	
bis(cyclopentadienyl)diphenyltitanium,	
bis(cyclopentadienyl)dineopentyltitanium,	
bis(cyclopentadienyl)dihydrotitanium,	
(cyclopentadienyl)(indenyl)titanium	dichloride,
(cyclopentadienyl)(fluorenyl)titanium	dichloride, etc.

(2) Transition metal compounds having two conjugated, 5-membered cyclic ligands, in which the two ligands are crosslinked with an alkylene group, such as methylenebis(indenyl)titanium dichloride, ethylenebis(indenyl)titanium dichloride, methylenebis(indenyl)titanium chlorohydride, ethylenebis(indenyl)methyltitanium chloride, ethylenebis(indenyl)methoxychlorotitanium, ethylenebis(indenyl)titanium diethoxide,

ethylenebis(indenyl)dimethyltitanium, ethylenebis(4,5,6,7-tetrahydroindenyl)titanium dichloride, ethylenebis(2-methylindenyl)titanium dichloride, ethylenebis(2,4-dimethylindenyl)titanium dichloride, ethylenebis(2-methyl-4-trimethylsilylindenyl)titanium dichloride, ethylenebis(2,4-dimethyl-5,6,7-trihydroindenyl)titanium dichloride, ethylene(2,4-dimethylcyclopentadienyl)(3',5'-dimethylcyclopentadienyl)titanium dichloride, ethylene(2-methyl-4-t-butylcyclopentadienyl)(3'-t-butyl-5'-methylcyclopentadienyl)titanium dichloride, ethylene(2,3,5-trimethylcyclopentadienyl)(2',4',5'-trimethylcyclopentadienyl)titanium dichloride, isopropylidenebis(2-methylindenyl)titanium dichloride, isopropylidenebis(indenyl)titanium dichloride, isopropylidenebis(2,4-dimethylindenyl)titanium dichloride, isopropylidene(2,4-dimethylcyclopentadienyl)(3',5'-dimethylcyclopentadienyl)titanium dichloride, isopropylidene(2-methyl-4-t-butylcyclopentadienyl)(3'-t-butyl-5'-methylcyclopentadienyl)titanium dichloride, methylene(cyclopentadienyl)(3,4-dimethylcyclopentadienyl)titanium dichloride, methylene(cyclopentadienyl)(3,4-dimethylcyclopentadienyl)titanium chlorohydrate, methylene(cyclopentadienyl)(3,4-dimethylcyclopentadienyl)dimethyltitanium,

methylene(cyclopentadienyl)(3,4-
 dimethylcyclopentadienyl)diphenyltitanium,
 methylene(cyclopentadienyl)(trimethylcyclopentadienyl)-
 titanium dichloride,
 methylene(cyclopentadienyl)(tetramethylcyclopentadienyl)-
 titanium dichloride, isopropylidene(cyclopentadienyl)(3,4-
 dimethylcyclopentadienyl)titanium dichloride,
 isopropylidene(cyclopentadienyl)(2,3,4,5-
 tetramethylcyclopentadienyl)titanium dichloride,
 isopropylidene(cyclopentadienyl)(3-methylindenyl)titanium
 dichloride,
 isopropylidene(cyclopentadienyl)(fluorenyl)titanium
 dichloride, isopropylidene(2-
 methylcyclopentadienyl)(fluorenyl)titanium dichloride,
 isopropylidene(2,5-dimethylcyclopentadienyl)(3,4-
 dimethylcyclopentadienyl)titanium dichloride,
 isopropylidene(2,5-
 dimethylcyclopentadienyl)(fluorenyl)titanium dichloride,
 ethylene(cyclopentadienyl)(3,5-
 dimethylcyclopentadienyl)titanium dichloride,
 ethylene(cyclopentadienyl)(fluorenyl)titanium dichloride,
 ethylene(2,5-dimethylcyclopentadienyl)(fluorenyl)titanium
 dichloride, ethylene(2,5-
 diethylcyclopentadienyl)(fluorenyl)titanium dichloride,
 diphenylmethylene(cyclopentadienyl)(3,4-

diethylcyclopentadienyl)titanium dichloride,
 diphenylmethylene(cyclopentadienyl)(3,4-
 diethylcyclopentadienyl)titanium dichloride,
 cyclohexylidene(cyclopentadienyl)(fluorenyl)titanium
 dichloride, cyclohexylidene(2,5-
 dimethylcyclopentadienyl)(3',4'-
 dimethylcyclopentadienyl)titanium dichloride, etc.

(3) Transition metal compounds having two silylene-
 crosslinked, conjugated, 5-membered cyclic ligands, such as
 dimethylsilylenebis(indenyl)titanium dichloride,
 dimethylsilylenebis(4,5,6,7-tetrahydroindenyl)titanium
 dichloride, dimethylsilylenebis(2-methylindenyl)titanium
 dichloride, dimethylsilylenebis(2,4-
 dimethylindenyl)titanium dichloride,
 dimethylsilylenebis(2,4-dimethylcyclopentadienyl)(3',5'-
 dimethylcyclopentadienyl)titanium dichloride,
 phenylmethylsilylenebis(indenyl)titanium dichloride,
 phenylmethylsilylenebis(4,5,6,7-tetrahydroindenyl)titanium
 dichloride, phenylmethylsilylenebis(2,4-
 dimethylindenyl)titanium dichloride,
 phenylmethylsilylene(2,4-dimethylcyclopentadienyl)(3',5'-
 dimethylcyclopentadienyl)titanium dichloride,
 phenylmethylsilylene(2,3,5-
 trimethylcyclopentadienyl)(2',4',5'-
 trimethylcyclopentadienyl)titanium dichloride,

phenylmethylsilylenebis(tetramethylcyclopentadienyl)-
 titanium dichloride, diphenylsilylenebis(2,4-
 dimethylindenyl)titanium dichloride,
 diphenylsilylenebis(indenyl)titanium dichloride,
 diphenylsilylenebis(2-methylindenyl)titanium dichloride,
 tetramethyldisilylenebis(indenyl)titanium dichloride,
 tetramethyldisilylenebis(cyclopentadienyl)titanium
 dichloride, tetramethyldisilylene(3-
 methylcyclopentadienyl)(indenyl)titanium dichloride,
 dimethylsilylene(cyclopentadienyl)(3,4-
 dimethylcyclopentadienyl)titanium dichloride,
 dimethylsilylene-
 (cyclopentadienyl)(trimethylcyclopentadienyl)titanium
 dichloride, dimethylsilylene-
 (cyclopentadienyl)(tetramethylcyclopentadienyl)titanium
 dichloride, dimethylsilylene(cyclopentadienyl)(3,4-
 diethylcyclopentadienyl)titanium dichloride,
 dimethylsilylene-
 (cyclopentadienyl)(triethylcyclopentadienyl)titanium
 dichloride, dimethylsilylene-
 (cyclopentadienyl)(tetraethylcyclopentadienyl)titanium
 dichloride,
 dimethylsilylene(cyclopentadienyl)(fluorenyl)titanium
 dichloride, dimethylsilylene(cyclopentadienyl)(2,7-di-t-
 butylfluorenyl)titanium dichloride, dimethylsilylene-

(cyclopentadienyl)(octahydrofluorenyl)titanium dichloride,
 dimethylsilylene(2-
 methylcyclopentadienyl)(fluorenyl)titanium dichloride,
 dimethylsilylene(2,5-
 dimethylcyclopentadienyl)(fluorenyl)titanium dichloride,
 dimethylsilylene(2-
 ethylcyclopentadienyl)(fluorenyl)titanium dichloride,
 dimethylsilylene(2,5-
 diethylcyclopentadienyl)(fluorenyl)titanium dichloride,
 diethylsilylene(2-methylcyclopentadienyl)(2',7'-di-t-
 butylfluorenyl)titanium dichloride, dimethylsilylene(2,5-
 dimethylcyclopentadienyl)(2',7'-di-t-
 butylfluorenyl)titanium dichloride, dimethylsilylene(2-
 ethylcyclopentadienyl)(2',7'-di-t-butylfluorenyl)titanium
 dichloride, dimethylsilylene(diethylcyclopentadienyl)(2,7-
 di-t-butylfluorenyl)titanium dichloride, dimethylsilylene-
 (methylcyclopentadienyl)(octahydrofluorenyl)titanium
 dichloride, dimethylsilylene-
 (dimethylcyclopentadienyl)(octahydrofluorenyl)titanium
 dichloride, dimethylsilylene-
 (ethylcyclopentadienyl)(octahydrofluorenyl)titanium
 dichloride, dimethylsilylene-
 (diethylcyclopentadienyl)(octahydrofluorenyl)titanium
 dichloride, etc.

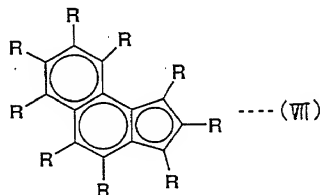
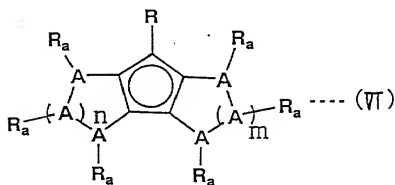
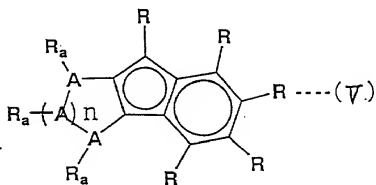
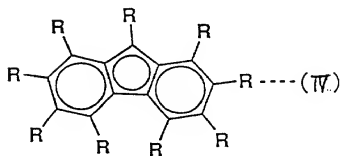
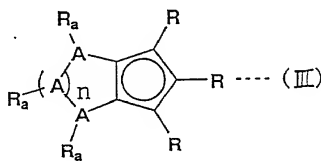
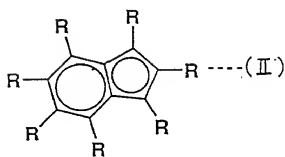
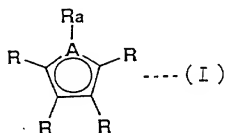
(4) Transition metal compounds having two conjugated, 5-membered cyclic ligands, in which the two ligands are crosslinked with a germanium-, aluminium-, boron-, phosphorus- or nitrogen-containing hydrocarbon group, such as dimethylgermylenebis(indenyl)titanium dichloride, dimethylgermylene(cyclopentadienyl)(fluorenyl)titanium dichloride, methylalumylenebis(indenyl)titanium dichloride, phenylamylenebis(indenyl)titanium dichloride, phenylphosphylenebis(indenyl)titanium dichloride, ethylborylenebis(indenyl)titanium dichloride, phenylamylenebis(indenyl)titanium dichloride, phenylamylene(cyclopentadienyl)(fluorenyl)titanium dichloride, etc.

(5) Transition metal compounds having one conjugated, 5-membered cyclic ligand, such as pentamethylcyclopentadienyl-bis(phenyl)aminotitanium dichloride, indenyl-bis(phenyl)aminotitanium dichloride, pentamethylcyclopentadienyl-bis(trimethylsilyl)aminotitanium dichloride, pentamethylcyclopentadienylphenoxytitanium dichloride, dimethylsilylene(tetramethylcyclopentadienyl)-phenylaminotitanium dichloride, dimethylsilylene(tetramethylcyclopentadienyl)-t-butylaminotitanium dichloride, dimethylsilylene(tetrahydroindenyl)decylaminotitanium

dichloride, dimethylsilylene(tetrahydroindenyl)-
 [bis(trimethylsilyl)amino]titanium dichloride,
 dimethylgermylene-
 (tetramethylcyclopentadienyl)phenylaminotitanium
 dichloride, pentamethylcyclopentadienyltitanium
 trimethoxide, pentamethylcyclopentadienyltitanium
 trichloride, (t-butylamido)(tetramethyl- η^5 -
 cyclopentadienyl)silane-titaniumdimethyl, (t-
 butylamido)(tetramethyl- η^5 -cyclopentadienyl)-1,2-ethane-
 diyltitanium dichloride, (methylamido)(tetramethyl- η^5 -
 cyclopentadienyl)-1,2-ethane-diyltitanium dichloride,
 (ethylamido)(tetramethyl- η^5 -cyclopentadienyl)-
 methylenetitanium dichloride, (t-butylamido)dimethyl-
 (tetramethyl- η^5 -cyclopentadienyl)silane-titaniumdichloride,
 (benzylamido)dimethyl-(tetramethyl- η^5 -
 cyclopentadienyl)silane-titanium dichloride,
 (phenylphosphido)dimethyl-(tetramethyl- η^5 -
 cyclopentadienyl)silane-titaniumdibenzyl, etc.

(6) Transition metal compounds having two conjugated,
 5-membered cyclic ligands in which the ligands are double-
 crosslinked, such as (1,1'-dimethylsilylene)(2,2'-
 isopropylene)bis(cyclopentadienyl)titanium dichloride,
 (1,1'-dimethylsilylene)(2,2'-dimethylsilylene)-
 bis(cyclopentadienyl)titanium dichloride, (1,1'-
 dimethylsilylene)(2,2'-isopropylidene)-

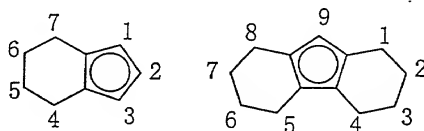
They are transition metal compounds of formula (I-4),
in which the group $(C_5H_{5-n}R^{11})_n$ is any of the following formulae
(I) to (VII):



wherein A represents an element of Group 13, 14, 15 or 16, and plural A's may be the same or different; R represents a hydrogen atom, a halogen atom, an aliphatic hydrocarbon group having from 1 to 30 carbon atoms, an aromatic hydrocarbon group having from 6 to 30 carbon atoms, an alkoxy group having from 1 to 30 carbon atoms, an aryloxy group having from 6 to 30 carbon atoms, a thioalkoxy group having from 1 to 30 carbon atoms, a thioaryloxy group having from 6 to 30 carbon atoms, an amino group, an amido group, a carboxyl group, or an alkylsilyl or alkylsilylalkyl group having from 3 to 30 carbon atoms, and R's may be the same or different, and may be optionally bonded to each other to form a cyclic structure; a represents 0, 1 or 2; and n and m each represent an integer of at least 1.

Specific examples of the group $(C_5H_{5-a}R^{11})_a$ are mentioned below.

In the indenyl derivatives and the fluorenyl derivatives, the position of each substituent is indicated by the following position numbers.



The group $(C_5H_{5-a}R^{11})_a$ includes, for example, a cyclopentadienyl group, a methylcyclopentadienyl group, a

1,2-dimethylcyclopentadienyl	group,	a	1,3-
dimethylcyclopentadienyl	group,	a	1,2,3-
trimethylcyclopentadienyl	group,	a	1,3,4-
trimethylcyclopentadienyl	group,	a	
tetramethylcyclopentadienyl	group,	a	
pentamethylcyclopentadienyl group, an ethylcyclopentadienyl			
group,	a	1,2-diethylcyclopentadienyl	group, a 1,3-
diethylcyclopentadienyl	group,	a	1,2,3-
triethylcyclopentadienyl	group,	a	1,3,4-
triethylcyclopentadienyl group, a tetraethylcyclopentadienyl			
group, a pentaethylcyclopentadienyl group, an indenyl group,			
a 1-methylindenyl group, a 1,2-dimethylindenyl group, a			
1,3-dimethylindenyl group, a 1,2,3-trimethylindenyl group, a			
2-methylindenyl group, a 1-ethylindenyl group, a 1-ethyl-			
2-methylindenyl group, a 1-ethyl-3-methylindenyl group, a			
1-ethyl-2,3-dimethylindenyl group, a 1,2-diethylindenyl			
group, a 1,3-diethylindenyl group, a 1,2,3-triethylindenyl			
group, a 2-ethylindenyl group, a 1-methyl-2-ethylindenyl			
group, a 1,3-dimethyl-2-ethylindenyl group, a 4,5,6,7-			
tetrahydroindenyl	group,	a	1-methyl-4,5,6,7-
tetrahydroindenyl	group,	a	1,2-dimethyl-4,5,6,7-
tetrahydroindenyl	group,	a	1,3-dimethyl-4,5,6,7-
tetrahydroindenyl	group,	a	1,2,3-trimethyl-4,5,6,7-
tetrahydroindenyl	group,	a	2-methyl-4,5,6,7-
tetrahydroindenyl group, a 1-ethyl-4,5,6,7-tetrahydroindenyl			

methylindenyltitaniumtrimethyl, 2-methylindenyltitanium
 trimethoxide, 1-methylindenyltitaniumtribenzyl, 1,2-
 dimethylindenyltitanium trichloride, 1,2-
 dimethylindenyltitaniumtrimethyl, 1,2-
 dimethylindenyltitanium trimethoxide, 1,2-
 dimethylindenyltitaniumtribenzyl, 1,3-
 dimethylindenyltitanium trichloride, 1,3-
 dimethylindenyltitaniumtrimethyl, 1,3-
 dimethylindenyltitanium trimethoxide, 1,3-
 dimethylindenyltitaniumtribenzyl, 1,2,3-
 trimethylindenyltitanium trichloride, 1,2,3-
 trimethylindenyltitaniumtrimethyl, 1,2,3-
 trimethylindenyltitanium trimethoxide, 1,2,3-
 trimethylindenyltitaniumtribenzyl, 1,2,3,4,5,6,7-
 heptamethylindenyltitanium trichloride, 1,2,3,4,5,6,7-
 heptamethylindenyltitaniumtrimethyl, 1,2,3,4,5,6,7-
 heptamethylindenyltitanium trimethoxide, 1,2,3,4,5,6,7-
 heptamethylindenyltitaniumtribenzyl, 4,5,6,7-
 tetrahydroindenyltitanium trichloride, 4,5,6,7-
 tetrahydroindenyltitaniumtrimethyl, 4,5,6,7-
 tetrahydroindenyltitanium trimethoxide, 4,5,6,7-
 tetrahydroindenyltitaniumtribenzyl, 1-methyl-4,5,6,7-
 tetrahydroindenyltitanium trichloride, 1-methyl-4,5,6,7-
 tetrahydroindenyltitaniumtrimethyl, 1-methyl-4,5,6,7-
 tetrahydroindenyltitanium trimethoxide, 1-methyl-4,5,6,7-

4,5,6,7-tetrahydroindenyltitanium trimethoxide, 1,3-
 diethyl-4,5,6,7-tetrahydroindenyltitaniumtribenzyl, 1,3-
 diethyl-2-methyl-4,5,6,7-tetrahydroindenyltitanium
 trichloride, 1,3-diethyl-2-methyl-4,5,6,7-
 tetrahydroindenyltitaniumtrimethyl, 1,3-diethyl-2-methyl-
 4,5,6,7-tetrahydroindenyltitanium trimethoxide, 1,3-
 diethyl-2-methyl-4,5,6,7-
 tetrahydroindenyltitaniumtribenzyl, 1,2,3-triethyl-
 4,5,6,7-tetrahydroindenyltitanium trichloride, 1,2,3-
 triethyl-4,5,6,7-tetrahydroindenyltitaniumtrimethyl,
 1,2,3-triethyl-4,5,6,7-tetrahydroindenyltitanium
 trimethoxide, 1,2,3-triethyl-4,5,6,7-
 tetrahydroindenyltitaniumtribenzyl, 2-ethyl-4,5,6,7-
 tetrahydroindenyltitanium trichloride, 2-ethyl-4,5,6,7-
 tetrahydroindenyltitaniumtrimethyl, 2-ethyl-4,5,6,7-
 tetrahydroindenyltitanium trimethoxide, 2-ethyl-4,5,6,7-
 tetrahydroindenyltitaniumtribenzyl, 1-methyl-2-ethyl-
 4,5,6,7-tetrahydroindenyltitanium trichloride, 1-methyl-2-
 ethyl-4,5,6,7-tetrahydroindenyltitaniumtrimethyl, 1-
 methyl-2-ethyl-4,5,6,7-tetrahydroindenyltitanium
 trimethoxide, 1-methyl-2-ethyl-4,5,6,7-
 tetrahydroindenyltitaniumtribenzyl, 1,3-dimethyl-2-ethyl-
 4,5,6,7-tetrahydroindenyltitanium trichloride, 1,3-
 dimethyl-2-ethyl-4,5,6,7-
 tetrahydroindenyltitaniumtrimethyl, 1,3-dimethyl-2-ethyl-

4,5,6,7-tetrahydroindenyltitanium trimethoxide, 1,3-dimethyl-2-ethyl-4,5,6,7-tetrahydroindenyltitaniumtribenzyl, 1,2,3,4-tetrahydrofluorenyltitanium trichloride, 1,2,3,4-tetrahydrofluorenyltitaniumtrimethyl, 1,2,3,4-tetrahydrofluorenyltitanium trimethoxide, 1,2,3,4-tetrahydrofluorenyltitaniumtribenzyl, 9-methyl-1,2,3,4-tetrahydrofluorenyltitanium trichloride, 9-methyl-1,2,3,4-tetrahydrofluorenyltitaniumtrimethyl, 9-methyl-1,2,3,4-tetrahydrofluorenyltitanium trimethoxide, 9-methyl-1,2,3,4-tetrahydrofluorenyltitaniumtribenzyl, 9-ethyl-1,2,3,4-tetrahydrofluorenyltitanium trichloride, 9-ethyl-1,2,3,4-tetrahydrofluorenyltitaniumtrimethyl, 9-ethyl-1,2,3,4-tetrahydrofluorenyltitanium trimethoxide, 9-ethyl-1,2,3,4-tetrahydrofluorenyltitaniumtribenzyl, 1,2,3,4,5,6,7,8-octahydrofluorenyltitanium trichloride, 1,2,3,4,5,6,7,8-octahydrofluorenyltitaniumtrimethyl, 1,2,3,4,5,6,7,8-octahydrofluorenyltitanium trimethoxide, 1,2,3,4,5,6,7,8-octahydrofluorenyltitaniumtribenzyl, 9-methyl-1,2,3,4,5,6,7,8-octahydrofluorenyltitanium trichloride, 9-methyl-1,2,3,4,5,6,7,8-octahydrofluorenyltitaniumtrimethyl, 9-methyl-1,2,3,4,5,6,7,8-octahydrofluorenyltitanium trimethoxide, 9-methyl-1,2,3,4,5,6,7,8-octahydrofluorenyltitanium trichloride, 9-ethyl-1,2,3,4,5,6,7,8-

octahydrofluorenyltitaniumtrimethyl, 9-ethyl-
 1,2,3,4,5,6,7,8-octahydrofluorenyltitanium trimethoxide,
 9-ethyl-1,2,3,4,5,6,7,8-
 octahydrofluorenyltitaniumtribenzyl, etc.; as well as their
 derivatives to be produced by substituting the titanium element
 in those compounds with zirconium or hafnium, or with any other
 element of different Groups, and also their analogues having
 a transition metal element of lanthanides. However, these
 are not limitative.

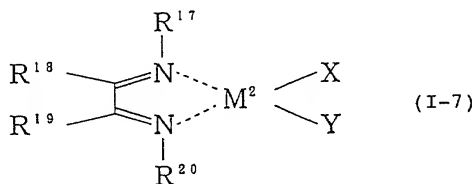
(III) Specific Examples of the transition metal
 compounds of formula (I-5) include tetramethyltitanium,
 tetrabenzyltitanium, tetraethyltitanium,
 tetraphenyltitanium, tetramethoxytitanium,
 tetraethoxytitanium, tetrephenoxytitanium,
 tetra(dimethylamino)titanium, tetra(diethylamino)titanium,
 tetra(diphenylamino)titanium; bis(phenoxy)titanium
 compounds described in Macromolecules, 1997, 30, 1562-1569,
 in Journal of Organometallic Chemistry, 514 (1996), 213-217,
 etc.; diamidotitanium compounds described in Macromolecules,
 1996, 29, 5241-5243, in Organometallics, 1997, 16, 1491-1496,
 etc.; their derivatives to be produced by substituting the
 titanium element in those compounds with zirconium or hafnium,
 or with any other element of different Groups, and also their
 analogues having a transition metal element of lanthanides.

(IV) In the transition metal compounds of formula (I-6), M^2 represents a transition metal of Groups 8 to 10 of the Periodic Table, concretely including iron, cobalt, nickel, palladium, platinum, etc. Of those, preferred are nickel and palladium. L^1 and L^2 each represents a coordination-bonding ligand; and X^1 and Y^1 each represent a covalent-bonding or ionic-bonding ligand. As mentioned hereinabove, X^1 and Y^1 include, for example, a hydrogen atom, a halogen atom, a hydrocarbon group having from 1 to 20, preferably from 1 to 10 carbon atoms, an alkoxy group having from 1 to 20, preferably from 1 to 10 carbon atoms, an amino group, a phosphorus-containing hydrocarbon group having from 1 to 20, preferably from 1 to 12 carbon atoms (e.g., a diphenylphosphine group, etc.), a silicon-containing hydrocarbon group having from 1 to 20, preferably from 1 to 12 carbon atoms (e.g., a trimethylsilyl group, etc.), and a boron compound residue having a hydrocarbon group with from 1 to 20, preferably from 1 to 12 carbon atoms or having halogens (e.g., $B(C_6H_5)_4$, BF_4). Of those, preferred are halogen atoms and hydrocarbon groups. X^1 and Y^1 may be the same or different ones. Specific examples of L^1 and L^2 include triphenylphosphine, acetonitrile, benzonitrile, 1,2-bisdiphenylphosphinoethane, 1,3-bisdiphenylphosphinopropane, 1,1'-bisdiphenylphosphinoferrocene, cyclooctadiene, pyridine,

bistrimethylsilylaminobistrimethylsilyliminophosphorane,
etc.

L^1 , L^2 , X^1 and Y^1 may be bonded to each other to form a cyclic structure.

The compounds of transition metals of Groups 8 to 10 of the Periodic Table preferably have a diimine compound as the ligand, including, for example, complex compounds of a general formula (I-7):



wherein R^{17} and R^{20} each independently represent an aliphatic hydrocarbon group having from 1 to 20 carbon atoms, or an aromatic group having a hydrocarbon group on the ring and having from 7 to 20 carbon atoms in total; R^{18} and R^{19} each independently represent a hydrogen atom, or a hydrocarbon group having from 1 to 20 carbon atoms, and R^{18} and R^{19} may be bonded to each other to form a ring; X and Y each independently represent a hydrogen atom, or a hydrocarbon group having from 1 to 20 carbon atoms; and M^2 represents a transition metal of Groups 8 to 10 of the Periodic Table.

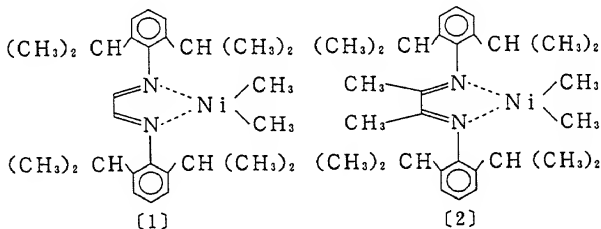
In formula (I-7), the aliphatic hydrocarbon group having from 1 to 20 carbon atoms for R^{17} and R^{20} may be a linear

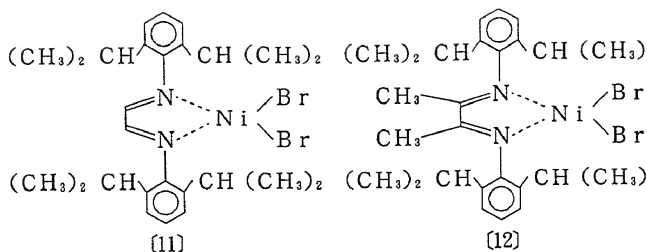
are those of the C1-20 aliphatic hydrocarbon group mentioned hereinabove for R^{17} and R^{20} . The aryl group having from 6 to 20 carbon atoms includes, for example, a phenyl group, a tolyl group, a xylyl group, a naphthyl group, a methylnaphthyl group, etc.; and the aralkyl group having from 7 to 20 carbon atoms includes, for example, a benzyl group, a phenethyl group, etc. R^{17} and R^{18} may be the same or different, and may be bonded to each other to form a ring.

For examples of the hydrocarbon group having from 1 to 20 carbon atoms for X and Y, referred to are those of the C1-20 hydrocarbon group mentioned hereinabove for R^{18} and R^{19} . For X and Y, especially preferred is a methyl group. X and Y may be the same or different.

The transition metal of Groups 8 to 10 of the Periodic Table for M^2 includes, for example, nickel, palladium, platinum, iron, cobalt, rhodium, ruthenium, etc. Preferred are nickel and palladium.

Specific examples of the complex compounds of formula (I-7) are compounds of the following formulae [1], [2], [3], [4], [5], [6], [7], [8], [9], [10], [11] and [12].





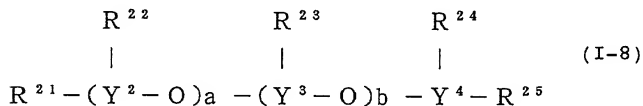
Specific examples of the transition metal compounds of formula (I-7) include dibromobistriphenylphosphine nickel, dichlorobistriphenylphosphine nickel, dibromodiacetonitrile nickel, dibromodibenzonitrile nickel, dibromo(1,2-bisdiphenylphosphinoethane) nickel, dibromo(1,3-bisdiphenylphosphinopropane) nickel, dibromo(1,1'-diphenylbisphosphinoferrocene) nickel, dimethylbisdiphenylphosphine nickel, dimethyl(1,2-bisdiphenylphosphinoethane) nickel, methyl(1,2-bisdiphenylphosphinoethane)nickel tetrafluoroborate, (2-diphenylphosphino-1-phenylethyleneoxy)phenylpyridine nickel, dichlorobistriphenylphosphine palladium, dichlorodibenzonitrile palladium, dichlorodiacetonitrile palladium, dichloro(1,2-bisdiphenylphosphinoethane) palladium, bistrisphenylphosphinopalladium bistetrafluoroborate, bis(2,2'-bipyridino)methyliron tetrafluoroborate etherate, etc.

Of those, preferred are cationic complexes such as methyl(1,2-bisdiphenylphosphinoethano)nickel tetrafluoroborate, bistrisphenylphosphinopalladium bistetrafluoroborate, and bis(2,2'-bipyridino)methyliron tetrafluoroborate etherate.

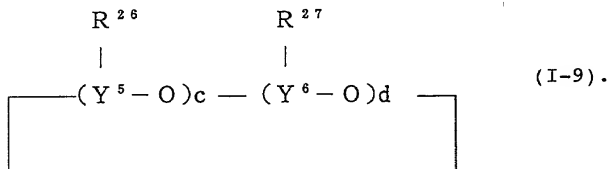
In the invention, one or more of the complex compounds noted above may be used either singly or as combined.

(B) Oxygen-containing compound:

Herein used are oxygen-containing compounds of a general formula (I-8):



and/or those of a general formula (I-9):



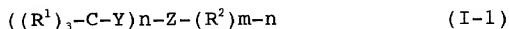
In formulae (I-8) and (I-9), R^{21} to R^{27} each represent an alkyl group having from 1 to 8 carbon atoms, concretely

including a methyl group, an ethyl group, an n-propyl group, an isopropyl group, all types of butyl groups, all types of pentyl groups, all types of hexyl group, all types of heptyl groups, and all types of octyl groups. R^{21} to R^{27} may be the same or different; and R^{26} and R^{27} may be the same or different. Y^2 to Y^6 each represent an element of Group 13 of the Periodic Table, concretely including B, Al, Ga, In and Tl. Of these, preferred are B and Al. Y^2 to Y^4 may be the same or different; and Y^5 and Y^6 may be the same or different. a to d each indicates a number of from 0 to 50, but (a + b) and (c + d) each must be at least 1. a to d each preferably falls between 1 and 20, more preferably between 1 and 10, even more preferably between 1 and 5.

As the oxygen-containing compound for the catalyst component, preferred are alkylaluminoxanes. Preferred examples of the compound are methylaluminoxane and isobutylaluminoxane.

Component (C):

This is a compound of the following general formula (I-1):



wherein R^1 represents an aromatic hydrocarbon group having from 6 to 30 carbon atoms, an alkoxy group having from 1 to 30 carbon atoms, an aryloxy group having from 6 to 30 carbon atoms, a thioalkoxy group having from 1 to 30 carbon atoms, a thioaryloxy

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group having from 6 to 30 carbon atoms, an amino group, an amido group, or a carboxyl group, R^1 's may be the same or different, and R^1 's may be optionally bonded to each other to form a cyclic structure; Y represents an element of Group 16; Z represents a metal element of Groups 2 to 13; R^2 represents a hydrocarbon group; m is an integer, indicating the valency of the metal element Z; and n is an integer of from 1 to (m-1),

Especially preferred are the following compounds: (1) Y is oxygen and Z is aluminium; (2) at least one of three R^1 's is an aromatic hydrocarbon group having from 6 to 30 carbon atoms; (3) three R^1 's are all hydrocarbon groups each having at least one carbon atom; (4) three R^1 's are all hydrocarbon groups each having from 4 to 30 carbon atoms; (5) three R^1 's are all aromatic hydrocarbon groups each having from 6 to 30 carbon atoms, preferably phenyl groups; (6) R^2 is an alkyl group having at least 2 carbon atoms.

Concretely, more preferred those where R^1 's are all phenyl groups, Y is oxygen, Z is aluminium, $n = 1$, and R^2 is an isobutyl group.

The compounds for the component (C) are not specifically defined for their production method, so far as they have the structure of the formula mentioned above, but preferred are those produced by reacting <1> a compound of a general formula, $(R^1)_3-C-OR^3$ or <1> at least one selected from compounds of a general formula, $(R^1)_3-C-OR^3$, R^4-CO-R^5 or $R^6-CO-OR^7$, with <2> a

compound of a general formula, $Z(R^2)_m$. (In these formulae, R^1 represents an aromatic hydrocarbon group having from 6 to 30 carbon atoms, an alkoxy group having from 1 to 30 carbon atoms, an aryloxy group having from 6 to 30 carbon atoms, a thioalkoxy group having from 1 to 30 carbon atoms, a thioaryloxy group having from 6 to 30 carbon atoms, an amino group, an amido group, or a carboxyl group, R^1 's may be the same or different, and R^1 's may be optionally bonded to each other to form a cyclic structure; R^3 , R^4 , R^5 , R^6 and R^7 each represent a hydrogen atom, a halogen atom, an aliphatic hydrocarbon group having from 1 to 30 carbon atoms, an aromatic hydrocarbon group having from 6 to 30 carbon atoms, an alkoxy group having from 1 to 30 carbon atoms, an aryloxy group having from 6 to 30 carbon atoms, a thioalkoxy group having from 1 to 30 carbon atoms, a thioaryloxy group having from 6 to 30 carbon atoms, an amino group, an amido group, or a carboxyl group, and R^3 , R^4 , R^5 , R^6 and R^7 may be the same or different; Z represents a metal element of Groups 2 to 13; m is an integer, indicating the valency of the metal element Z ; and R^2 represents a hydrocarbon group.

Concretely, they are reaction products of at least one selected from alcohols, ethers, aldehydes, ketones, carboxylic acids and carboxylates, with an aluminium compound. Preferred are reaction products of alcohols with an aluminium compound. For these, preferred are the following: (1) at least one of three R^1 's for $(R^1)_3$ is an aromatic hydrocarbon

In place of using the compound of the above-mentioned general formula as the component (C), a compound of the following (C1) and a compound of the following (C2) may be directly added to the site of catalyst production or to the site of polymerization to form the component (C) in situ. In this case, the catalyst components are the transition metal

compound (A), the oxygen-containing compound (B), the compounds (C1) and (C2), and optionally an alkylating agent (D).

(C1) is a compound of a general formula, $(R^1)_3-C-OR^3$, or at least one selected from compounds of a general formula, $(R^1)_3-C-OR^3$, R^4-CO-R^5 or $R^6-CO-OR^7$; and (C2) is a compound of a general formula, $Z(R^2)_m$.

In these formulae, R^1 represents an aromatic hydrocarbon group having from 6 to 30 carbon atoms, an alkoxy group having from 1 to 30 carbon atoms, an aryloxy group having from 6 to 30 carbon atoms, a thioalkoxy group having from 1 to 30 carbon atoms, a thioaryloxy group having from 6 to 30 carbon atoms, an amino group, an amido group, or a carboxyl group, R^1 's may be the same or different, and R^1 's may be optionally bonded to each other to form a cyclic structure; R^3 , R^4 , R^5 , R^6 and R^7 each represent a hydrogen atom, a halogen atom, an aliphatic hydrocarbon group having from 1 to 30 carbon atoms, an aromatic hydrocarbon group having from 6 to 30 carbon atoms, an alkoxy group having from 1 to 30 carbon atoms, an aryloxy group having from 6 to 30 carbon atoms, a thioalkoxy group having from 1 to 30 carbon atoms, a thioaryloxy group having from 6 to 30 carbon atoms, an amino group, an amido group, or a carboxyl group, and R^3 , R^4 , R^5 , R^6 and R^7 may be the same or different; Z represents a metal element of Groups 2 to 13; m is an integer,

indicating the valency of the metal element Z; and R² represents a hydrocarbon group.

Concretely, (C1) is at least one selected from alcohols, ethers, aldehydes, ketones, carboxylic acids and carboxylates, preferably from alcohols; and (C2) is an aluminium compound. For these, preferred are the following: (1) at least one of three R¹'s for (R¹), is an aromatic hydrocarbon group having from 6 to 30 carbon atoms; (2) three R¹'s for (R¹), are all hydrocarbon groups each having at least one carbon atom; (3) three R¹'s for (R¹), are all hydrocarbon groups each having from 4 to 30 carbon atoms; (4) three R¹'s for (R¹), are all aromatic hydrocarbon groups each having from 6 to 30 carbon atoms, preferably phenyl groups; (5) R² is an alkyl group having at least 2 carbon atoms. More concretely, the most preferred combination is (C1) of triphenylmethyl alcohol and (C2) of triisobutylaluminium.

(D) Alkylating agent:

The catalyst of the invention for production of olefinic polymers optionally contains an alkylating agent. Various types of alkylating agents are known and are usable in the invention, including, for example, alkyl group-having aluminium compounds of a general formula (I-12):



wherein R²⁸ and R²⁹ each represent an alkyl group having from 1 to 8, preferably from 1 to 4 carbon atoms; X represents

a hydrogen atom or a halogen atom; $0 < m \leq 3$, but preferably m is 2 or 3, most preferably 3; $0 \leq n < 3$, but preferably n is 0 or 1;

alkyl group-having magnesium compounds of a general formula (I-13):



wherein R^{28} has the same meaning as above; and alkyl group-having zinc compounds of a general formula (I-14):



wherein R^{28} has the same meaning as above.

Of these alkyl group-having compounds, preferred are alkyl group-having aluminium compounds; and more preferred are trialkylaluminium compound and dialkylaluminium compounds. Concretely, they include trialkylaluminiums such as trimethylaluminium, triethylaluminium, tri-n-propylaluminium, triisopropylaluminium, tri-n-butylaluminium, triisobutylaluminium, tri-t-butylaluminium, etc.; dialkylaluminium halides such as dimethylaluminium chloride, diethylaluminium chloride, di-n-propylaluminium chloride, diisopropylaluminium chloride, di-n-butylaluminium chloride, diisobutylaluminium chloride, di-t-butylaluminium chloride, etc.; dialkylaluminium alkoxides such as dimethylaluminium methoxide, dimethylaluminium ethoxide, etc.; dialkylaluminium hydrides such as dimethylaluminium

hydride, diethylaluminium hydride, diisobutylaluminium hydride, etc. Further mentioned are dialkylmagnesiums such as dimethylmagnesium, diethylmagnesium, di-n-propylmagnesium, diisopropylmagnesium, etc.; dialkylzincs such as dimethylzinc, diethylzinc, di-n-propylethylzinc, diisopropylzinc, etc.

2. Method for producing catalyst:

(1) Order of contacting constituent components:

To produce the catalyst of the invention, the order of contacting the constituent components with each other is not specifically defined. For example, the components may be contacted with each other in the following manner.

(i) For producing the catalyst comprising the component (A), the component (B) and the component (C), for example, employable is <1> a method of first contacting the component (A) with the component (B), followed by further contacting it with the component (C); <2> a method of first contacting the component (A) with the component (C) followed by further contacting it with the component (B); <3> a method of first contacting the component (B) with the component (C) followed by further contacting it with the component (A); or <4> a method of contacting the three components all together.

When the catalyst contains the optional component (D), the order of contacting the component (D) with the other components is not specifically defined. For example, in the process of producing the catalyst, the component (A) may be

ratio of the component (A) to the component (C) may fall between 1/0.5 and 1/1,000, but preferably between 1/0.8 and 1/100, more preferably between 1/1 and 1/100, in terms of the molar ratio to the aluminium atom in (C). Regarding the ratio by mol of the component (A) to the component (D), referred to is a case where an aluminium compound is used as the component (D). In that case, the molar ratio of the component (A) to the component (D) may fall between 1/0.5 and 1/1,000, but preferably between 1/1 and 1/100, in terms of the molar ratio to the aluminium atom in (D).

<3> Where the catalyst contains a combination of the component (C1) and the component (C2), but not the component (C), the molar ratio of (C1) to (C2) may fall between 1/0.1 and 1/50, precisely between 1/0.1 and 1/10, or between 1/0.5 and 1/20, or between 1/0.8 and 1/10, or between 1/0.5 and 1/2, or between 1/0.8 and 1/1.2, but most preferably between 1/0.8 and 1/10.

Regarding the ratio by mol of the component (A) to the component (C2), referred to is a case where an aluminium compound is used as the component (C2). In that case, the molar ratio of the component (A) to the component (C2) may fall between 1/0.5 and 1/1,000, but preferably between 1/1 and 1/100, in terms of the molar ratio to the aluminium atom in (C2). Regarding the blend ratio of the component (D), the same as in the case <2> shall apply also to the case <3>.

(3) Condition for contacting constituent components:

To produce the catalyst, the constituent components may be contacted with each other in an inert atmosphere of nitrogen or the like, at a temperature not higher than the temperature at which the catalyst is used for polymerization. As the case may be, they may be contacted with each other at a temperature falling between -30 and 200°C .

(4) In the invention, the components constituting the catalyst, especially the component (B) may be carried on a suitable carrier. Any types of carriers are usable with no specific limitation, but preferred are inorganic carriers such as inorganic oxides, etc. Concretely, the inorganic oxides include SiO_2 , Al_2O_3 , TiO_2 , Fe_2O_3 , B_2O_3 , CaO , ZnO , BaO , ThO_2 , silica-alumina, zeolite, ferrite, glass fibers, etc. Of those, especially preferred are SiO_2 and Al_2O_3 . The inorganic oxide carriers may contain a small amount of carbonates, nitrates, sulfates, etc. For their properties, the carriers are not specifically defined. Generally, the carriers have a mean particle size of from 1 to $300\text{ }\mu\text{m}$, but preferably from 10 to $200\text{ }\mu\text{m}$, more preferably from 20 to $100\text{ }\mu\text{m}$.

II. Method for producing olefinic polymers:

1. Monomers to be polymerized:

In the method of the invention for producing olefinic polymers, used is the above-mentioned catalyst for polymerization of olefins. The catalyst is favorable to

homopolymerization of olefins and to copolymerization of olefins with other olefins (that is, copolymerization of different types of olefins).

Olefins for the method are not specifically defined, but preferred are α -olefins having from 2 to 20 carbon atoms. Especially preferred are ethylene and propylene.

They include α -olefins such as ethylene, propylene, 1-butene, 3-methyl-1-butene, 4-methyl-1-butene, 4-phenyl-1-butene, 1-pentene, 3-methyl-1-pentene, 4-methyl-1-pentene, 3,3-dimethyl-1-pentene, 3,4-dimethyl-1-pentene, 4,4-dimethyl-1-pentene, 1-hexene, 4-methyl-1-hexene, 5-methyl-1-hexene, 6-phenyl-1-hexene, 1-octene, 1-decene, 1-dodecene, 1-tetradecene, 1-hexadecene, 1-octadecene, 1-eicosene, vinylcyclohexane, etc.; halogen-substituted α -olefins such as hexafluoropropene, tetrafluoroethylene, 2-fluoropropene, fluoroethylene, 1,1-difluoroethylene, 3-fluoropropene, trifluoroethylene, 3,4-dichloro-1-butene, etc.; and cyclic olefins such as cyclopentene, cyclohexene, norbornene, 5-methylnorbornene, 5-ethylnorbornene, 5-propylnorbornene, 5,6-dimethylnorbornene, 5-benzylnorbornene, etc.

In the invention, one or more olefins such as those mentioned above may be (co)polymerized in any desired combination.

2. Polymerization condition:

In the invention, the polymerization catalyst may be

component (C) or with the components (C1) and (C2) in a different reactor, and the two blends in different reactors are mixed just before the start of polymerization of the monomers. Having been thus blended, the monomers begin to polymerize in the presence of the catalyst formed in situ.

For the polymerization, a solvent may be used. The solvent includes hydrocarbons and halogenohydrocarbons such as benzene, toluene, xylene, n-hexane, n-heptane, cyclohexane, methylene chloride, chloroform, 1,2-dichloroethane, chlorobenzene, etc. One or more of these compounds may be used either singly or as combined for the solvent. Depending on their type, the monomers to be polymerized could serve as a polymerization solvent.

The amount of the catalyst to be used for the polymerization is preferably so controlled that the component (A) may fall generally between 0.1 and 100 μ moles, but preferably between 0.5 and 25 μ moles, in one liter of the solvent, in view of the polymerization activity and the reactor efficiency.

Regarding the polymerization condition, the pressure may fall generally between normal pressure and 2000 kg/cm²G (196 MPaG). The reaction temperature may fall generally between -50 and 250°C. For controlling the molecular weight of the polymer to be produced, the type and the amount of the catalyst components to be used and the polymerization temperature shall be suitably controlled, or hydrogen may be

introduced into the polymerization system.

The invention is described in more detail with reference to the following Examples, which, however, are not intended to restrict the scope of the invention.

The stereospecificity [mmmm] of the polymer produced was determined as follows: A sample of the polymer was dissolved in a mixed solvent of 1,2,4-trichlorobenzene and heavy benzene (90/10, by volume), and subjected to a proton complete decoupling method for ^{13}C -NMR (using JEOL' LA-500) at 130°C. Based on the signals for the methyl group obtained in the method, the stereospecificity [mmmm] of the sample was determined. The stereospecificity was proposed by A. Zambelli et al. in *Macromolecules*, 6, 925 (1973), and it indicates the isotactic fraction [mmmm] in the pentad units of a polypropylene molecular chain as measured in ^{13}C nuclear magnetic resonance spectrometry. For the attribution of the peaks seen in the ^{13}C nuclear magnetic resonance spectrometry, referred to was the A. Zambelli et al's proposal in *Macromolecules*, 8, 687 (1975).

The molecular weight (M_w) of the polymer produced and the molecular weight distribution (M_w/M_n) thereof were measured through GPC.

[Example I-1]

(1) Preparation of component (C):

0.875 ml of 2 M triisobutylaluminium was added to a

toluene solution of 455 mg (1.75 mmols) of triphenylmethanol at -78°C , and stirred at room temperature for 24 hours. The concentration of the component (C) thus formed was 0.1 mols/liter.

Through $^1\text{H-NMR}$, the product was confirmed to have the component (C) formed therein.

(2) Preparation of silica-carried methylaluminoxane:

27.1 g of SiO_2 (Fuji Silicia's P-10) was put into a 500 ml Schlenk, and dried at 200°C under pressure for 4 hours. The baked SiO_2 weighed 25.9 g. The baked SiO_2 was put into 400 ml of toluene cooled at -78°C in a dry ice/methanol bath, and stirred. With stirring, 145.5 ml of a solution of methylaluminoxane/toluene (1.5 mols/liter) was dropwise added to the resulting suspension, which took 1 hour to the end. After left as such for 4 hours, this was heated from -78°C up to 20°C over a period of 6 hours, and then further left as it was for 4 hour. Next, this was heated from 20°C up to 80°C over a period of 1 hour, and then left at 80°C for 4 hours. Through the process, the reaction between silica and methylaluminoxane was completed. The resulting suspension was filtered at 60°C , and the solid residue was washed twice with 400 ml of toluene at 60°C . The thus-washed solid was dried at 60°C under reduced pressure for 4 hours, whereby was obtained 33.69 g of silica-carried methylaluminoxane in which the proportion of methylaluminoxane was 23.12 % by weight. N-heptane was added

to all the silica-carried methylaluminoxane to be 500 ml in total. The suspension thus prepared had a methylaluminoxane concentration of 0.27 mols/liter.

(3) Polymerization of propylene:

400 ml of heptane was put into a one-liter stainless autoclave, to which were added 0.5 mmols of triisobutylaluminium, 50 μ mol of the solution of the component (C) prepared in the step (1), 0.25 mmols, in terms of aluminium atom, of the silica-carried methylaluminoxane prepared in the step (2), and 1 μ mol of dimethylsilylenebis(2-methyl-4,5-benzoindenyl)zirconium dichloride after having been pre-contacted with each other in toluene for 5 minutes to form a catalyst. Propylene gas was introduced into the autoclave to have a total pressure of 0.785 MPaG (8.0 kg/cm²G). During its polymerization, propylene was continuously introduced into the autoclave via a pressure controlling unit so as to have all the time a constant pressure. In that condition, propylene was polymerized at 70°C for 60 minutes. The resulting product was dried under reduced pressure. Thus obtained, the propylene polymer weighed 33.2 g. Its limiting viscosity (at 135°C in tetralin), $[\eta]$, was 1.2; its weight-average molecular weight (Mw) was 1,070,000; its molecular weight distribution (Mw/Mn) was 2.07; and its stereospecificity was 97.2 %.

[Comparative Example I-1]

(1) Blending of component of catalyst for polymer production:

This is the same as in Example I-1 (2).

(2) Polymerization of olefin:

This is the same as in Example I-1, except for the step of Example I-1 (3). In this, the component (C) prepared in Example I-1 (1) was not added to the polymerization system.

Thus obtained, the propylene polymer weighed 16.0 g. Its limiting viscosity (at 135°C in tetralin), $[\eta]$, was 1.3; its weight-average molecular weight (M_w) was 1,100,000; its molecular weight distribution (M_w/M_n) was 2.04; and its stereospecificity was 96.8 %.

[Example I-2]

(1) Preparation of component (C):

0.875 ml of 2 M triisobutylaluminium was added to a toluene solution of 1.75 mmols of tricyclohexylmethanol (from Across) at -78°C, and stirred at room temperature for 24 hours. The concentration of the component (C) thus formed was 0.1 mols/liter.

Through $^1\text{H-NMR}$, the product was confirmed to have the component (C) formed therein.

(2) Polymerization of propylene:

400 ml of heptane was put into a one-liter stainless autoclave, to which were added 0.5 mmols of triisobutylaluminium, 50 μmols of the solution of the component (C) prepared in the step (1), 0.25 mmols, in terms of aluminium atom, of the silica-carried methylaluminoxane prepared in the

step of Example I-1 (2), and 1 μmol of dimethylsilylenebis(2-methyl-4,5-benzoindenyl)zirconium dichloride after having been pre-contacted with each other in toluene for 5 minutes to form a catalyst. Propylene gas was introduced into the autoclave to have a total pressure of 0.785 MPaG (8.0 kg/cm²G). During its polymerization, propylene was continuously introduced into the autoclave via a pressure controlling unit so as to have all the time a constant pressure. In that condition, propylene was polymerized at 70°C for 60 minutes. The resulting product was dried under reduced pressure. Thus obtained, the propylene polymer weighed 28.4 g. Its weight-average molecular weight (M_w) was 1,190,000; its molecular weight distribution (M_w/M_n) was 2.04; and its stereospecificity was 97.1 %.

[Example I-3]

(1) Polymerization of ethylene-1-octene:

360 ml of toluene, 40 ml of 1-octene, 0.5 ml of a toluene solution of 1 M triisobutylaluminium, 50 μmol s of the solution of the component (C) prepared in the step of Example I-1 (1), and 0.25 mmols, in terms of aluminium atom, of the silica-carried methylaluminoxane prepared in the step of Example I-1 (2) were put into a 1.6-liter stainless autoclave equipped with a catalyst feeder tube, in that order, and heated up to 70°C. Next, ethylene was introduced into the autoclave to have a pressure of 0.785 MPaG (8.0 kg/cm²G). Next, 1.0 μmol of

(t-butylamido)dimethyl(tetramethyl- η 5-cyclopentadienyl)silane-titanium dichloride dissolved in 20 ml of toluene was put into the autoclave via the catalyst feeder tube.

Ethylene was continuously introduced into the autoclave so as to have all the time the constant pressure of 0.785 MPaG (8.0 kg/cm²G), and polymerized for 60 minutes. Next, methanol was added to this to stop the polymerization. A large amount of methanol was added to this, and the polymer formed was separated through filtration, and then dried at 60°C under reduced pressure for 4 hours. Thus obtained, the ethylene-1-octene copolymer weighed 33.8 g. Its weight-average molecular weight (M_w) was 61,000 in terms of ethylene; and its molecular weight distribution (M_w/M_n) was 2.24.

The 1-octene content of the copolymer was 18.4 mol%.
[Comparative Example I-2]

The same process as in Example I-3 was repeated, except that the component (C) was not added to the polymerization system. As a result, herein obtained was 18.3 g of ethylene-1-octene copolymer. Its weight-average molecular weight (M_w) was 64,000 in terms of ethylene; and its molecular weight distribution (M_w/M_n) was 2.06.

The 1-octene content of the copolymer was 18.3 mol%.
[Example I-4]

The same process as in Example I-3 was repeated, except

that 2,2-t-butyl-4,4-methylthiobisphenoxytitanium dichloride was used in place of (t-butylamido)dimethyl(tetramethyl- η 5-cyclopentadienyl)silane-titanium dichloride. As a result, herein obtained was 29.3 g of ethylene-1-octene copolymer. Its weight-average molecular weight (M_w) was 31,000 in terms of ethylene; and its molecular weight distribution (M_w/M_n) was 2.24.

The 1-octene content of the copolymer was 18.4 mol%.
[Comparative Example I-3]

The same process as in Example I-4 was repeated, except that the component (C) was not added to the polymerization system. As a result, herein obtained was 19.4 g of ethylene-1-octene copolymer. Its weight-average molecular weight (M_w) was 34,000 in terms of ethylene; and its molecular weight distribution (M_w/M_n) was 2.06.

The 1-octene content of the copolymer was 18.3 mol%.

[Second aspect of the invention]

In this section, the second aspect of the invention will be simply referred to as "the invention".

I. Catalyst for polymerization of olefins:

1. Components of catalyst for polymerization of olefins:

The catalyst of the invention for polymerization of olefins comprises (A) a transition metal compound, (B) a

compound capable of reacting with a transition metal compound to form an ionic complex, and (C) a compound of formula (II-1) which will be described in detail hereinunder, and optionally (D) an alkylating agent. The constituent components are described below.

(1) (A) Transition metal compound:

For its details, the same as in the section of the first aspect shall directly apply also to this section.

(B) Compound capable of reacting with a transition metal compound to form an ionic complex:

The compound capable of reacting with a transition metal compound to form an ionic complex includes coordination complex compounds that comprise an anion with plural groups bonded to a metal, and a cation, and Lewis acids. Known are various coordination complex compounds that comprise an anion with plural groups bonded to a metal, and a cation. For example, compounds of the following general formula (II-2) or (II-3) are preferably used in the invention.



wherein L^{24} indicates M^{25} , $R^{51}R^{52}M^{26}$ or R^{53}_3C , which will be mentioned hereinafter; L^{23} represents a Lewis base; M^{23} and M^{24} each represent a metal selected from Group 5 to Group 15 of the Periodic Table; M^{25} represents a metal selected from Group 1, and Group 8 to Group 12 of the Periodic Table; M^{26}

represents a metal selected from Group 8 to Group 10 of the Periodic Table; X^{22} to X^n each represent a hydrogen atom, a dialkylamino group, an alkoxy group, an aryloxy group, an alkyl group having from 1 to 20 carbon atoms, an aryl group having from 6 to 20 carbon atoms, an alkylaryl group, an arylalkyl group, a substituted alkyl group, an organometalloid group, or a halogen atom; R^{51} and R^{52} each represent a cyclopentadienyl group, a substituted cyclopentadienyl group, an indenyl group, or a fluorenyl group; R^{53} represents an alkyl group; m indicates the valency of M^{23} or M^{24} , and represents an integer of from 1 to 7; n represents an integer of from 2 to 8; g indicates the ion valency of $L^{23}-H$ or L^{24} , and represents an integer of from 1 to 7; h represents an integer of 1 or more; and $i = h \times g / (n-m)$.

Specific examples of M^{23} and M^{24} include atoms of B, Al, Si, P, As, Sb, etc.; those of M^{25} include atoms of Ag, Cu, Na, Li, etc.; and those of M^{26} include atoms of Fe, Co, Ni, etc. Specific examples of X^{22} to X^n include a dimethylamino group, a diethylamino group, etc., as a dialkylamino group; a methoxy group, an ethoxy group, an n-butoxy group, etc., as an alkoxy group; a phenoxy group, a 2,6-dimethylphenoxy group, a naphthyloxy group, etc., as an aryloxy group; a methyl group, an ethyl group, an n-propyl group, an isopropyl group, an n-butyl group, an n-octyl group, a 2-ethylhexyl group, etc., as an alkyl group having from 1 to 20 carbon atoms; a phenyl

group, a p-tolyl group, a benzyl group, a pentafluorophenyl group, a 3,5-di(trifluoromethyl)phenyl group, a 4-tert-butylphenyl group, a 2,6-dimethylphenyl group, a 3,5-dimethylphenyl group, a 2,4-dimethylphenyl group, a 1,2-dimethylphenyl group, etc., as an aryl group having from 6 to 20 carbon atoms, an alkylaryl group or an arylalkyl group; F, Cl, Br, I, as a halogen; and a pentamethylantimonyl group, a trimethylsilyl group, a trimethylgermyl group, a diphenylarsenyl group, a dicyclohexylantimonyl group, a diphenylboryl group, etc., as an organometalloid group. Specific examples of the substituted cyclopentadienyl group for R⁵¹ and R⁵² include a methylcyclopentadienyl group, a butylcyclopentadienyl group, a pentamethylcyclopentadienyl group, etc.

In the invention, concretely, the anion with plural groups bonded to a metal includes $B(C_6F_5)_4^-$, $B(C_6HF_4)_4^-$, $B(C_6H_2F_3)_4^-$, $B(C_6H_3F_2)_4^-$, $B(C_6H_4F)_4^-$, $B[C_6(CF_3)F_4]_4^-$, $B(C_6H_5)_4^-$, PF_6^- , $P(C_6F_5)_6^-$, $Al(C_6HF_4)_4^-$, etc. The cation includes, for example, Cp_2Fe^+ , $(MeCp)_2Fe^+$, $(tBuCp)_2Fe^+$, $(Me_2Cp)_2Fe^+$, $(Me_3Cp)_2Fe^+$, $(Me_4Cp)_2Fe^+$, $(Me_5Cp)_2Fe^+$, Ag^+ , Na^+ , Li^+ , etc. The other cations include, for example, those from nitrogen-containing compounds, such as pyridinium, 2,4-dinitro-N,N-diethylanilinium, diphenylammonium, p-nitroanilinium, 2,5-dichloroanilinium, p-nitro-N,N-dimethylanilinium, quinolinium, N,N-dimethylanilinium,

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N,N-diethylanilinium, etc.; those from carbenium compounds such as triphenylcarbenium, tri(4-methylphenyl)carbenium, tri(4-methoxyphenyl)carbenium, etc.; alkylphosphonium ions such as CH_3PH_3^+ , $\text{C}_2\text{H}_5\text{PH}_3^+$, $\text{C}_3\text{H}_7\text{PH}_3^+$, $(\text{CH}_3)_2\text{PH}_2^+$, $(\text{C}_2\text{H}_5)_2\text{PH}_2^+$, $(\text{C}_3\text{H}_7)_2\text{PH}_2^+$, $(\text{CH}_3)_3\text{PH}^+$, $(\text{C}_2\text{H}_5)_3\text{PH}^+$, $(\text{C}_3\text{H}_7)_3\text{PH}^+$, $(\text{CF}_3)_3\text{PH}^+$, $(\text{CH}_3)_4\text{P}^+$, $(\text{C}_2\text{H}_5)_4\text{P}^+$, $(\text{C}_3\text{H}_7)_4\text{P}^+$, etc.; arylphosphonium ions such as $\text{C}_6\text{H}_5\text{PH}_3^+$, $(\text{C}_6\text{H}_5)_2\text{PH}_2^+$, $(\text{C}_6\text{H}_5)_3\text{PH}^+$, $(\text{C}_6\text{H}_5)_4\text{P}^+$, $(\text{C}_2\text{H}_5)_2(\text{C}_6\text{H}_5)\text{PH}^+$, $(\text{CH}_3)(\text{C}_6\text{H}_5)\text{PH}_2^+$, $(\text{CH}_3)_2(\text{C}_6\text{H}_5)\text{PH}^+$, $(\text{C}_2\text{H}_5)_2(\text{C}_6\text{H}_5)_2\text{P}^+$, etc.

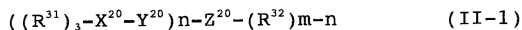
Of the compounds of formulae (II-2) and (II-3), concretely, the following are especially preferred. Preferred examples of the compounds of formula (II-2) include triethylammonium tetraphenylborate, tri(n-butyl)ammonium tetraphenylborate, trimethylammonium tetraphenylborate, triethylammonium tetrakis(pentafluorophenyl)borate, tri(n-butyl)ammonium tetrakis(pentafluorophenyl)borate, triethylammonium hexafluoroarsenate, pyridinium tetrakis(pentafluorophenyl)borate, pyrrolinium tetra(pentafluorophenyl)borate, N,N-dimethylanilinium tetrakis(pentafluorophenyl)borate, methyl diphenylammonium tetrakis(pentafluorophenyl)borate, etc. Preferred examples of the compounds of formula (II-3) include ferrocenium tetraphenylborate, dimethylferrocenium tetrakis(pentafluorophenyl)borate, ferrocenium tetrakis(pentafluorophenyl)borate, decamethylferrocenium

tetrakis(pentafluorophenyl)borate, acetylferrocenium
 tetrakis(pentafluorophenyl)borate, formylferrocenium
 tetrakis(pentafluorophenyl)borate, cyanoferrocenium
 tetrakis(pentafluorophenyl)borate, silver tetraphenylborate,
 silver tetrakis(pentafluorophenyl)borate, trityl
 tetraphenylborate, trityl tetrakis(pentafluorophenyl)borate,
 silver hexafluoroarsenate, silver hexafluoroantimonate,
 silver tetrafluoroborate, etc.

The Lewis acid includes, for example, $B(C_6F_5)_3$,
 $B(C_6HF_4)_3$, $B(H_2F_3)_3$, $B(C_6H_3F_2)_3$, $B(C_6H_4F)_3$, $B(C_6H_5)_3$, BF_3 ,
 $B[C_6(CF_3)F_4]_3$, PF_5 , $P(C_6F_5)_5$, $Al(C_6HF_4)_3$, etc.

(C) Component:

This is a compound of the following general formula
 (II-1):



wherein R^{31} represents a hydrogen atom, a halogen atom, an
 aliphatic hydrocarbon group having from 1 to 30 carbon atoms,
 an aromatic hydrocarbon group having from 6 to 30 carbon atoms,
 an alkoxy group having from 1 to 30 carbon atoms, an aryloxy
 group having from 6 to 30 carbon atoms, a thioalkoxy group
 having from 1 to 30 carbon atoms, a thioaryloxy group having
 from 6 to 30 carbon atoms, an amino group, an amido group, or
 a carboxyl group, R^{31} 's may be the same or different, and R^{31} 's
 may be optionally bonded to each other to form a cyclic
 structure; X^{20} represents an element of Group 14; Y^{20} represents

an element of Group 16; Z^{20} represents a metal element of Groups 2 to 13; R^{32} represents a hydrocarbon group; m is an integer, indicating the valency of the metal element; and n is an integer of from 1 to (m-1).

Especially preferred are the following compounds: (1) X^{20} is carbon, Y^{20} is oxygen, and Z^{20} is aluminium; (2) at least one of three R^{31} 's is an aromatic hydrocarbon group having from 6 to 30 carbon atoms; (3) three R^{31} 's are all hydrocarbon groups each having at least one carbon atom; (4) three R^{31} 's are all hydrocarbon groups each having from 4 to 30 carbon atoms; (5) three R^{31} 's are all aromatic hydrocarbon groups each having from 6 to 30 carbon atoms, preferably phenyl groups; (6) R^{32} is an alkyl group having at least 2 carbon atoms.

Concretely, more preferred those where R^{31} 's are all phenyl groups, X^{20} is carbon, Y^{20} is oxygen, Z^{20} is aluminium, $n = 1$, and R^{32} is an isobutyl group.

The compounds for the component (C) are not specifically defined for their production method, so far as they have the structure of the formula mentioned above, but preferred are those produced by reacting <1> a compound of a general formula, $(R^{31})_3-C-OR^{33}$ or <1> at least one selected from compounds of a general formula, $(R^{31})_3-C-OR^{33}$, $R^{34}-CO-R^{35}$ or $R^{36}-CO-OR^{37}$, with <2> a compound of a general formula, $Z(R^{32})_m$. (In these formulae, R^{31} represents a hydrogen atom, a halogen atom, an aliphatic hydrocarbon group having from 1 to 30 carbon atoms, an aromatic

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groups each having at least one carbon atom; (3) three R^{31} 's are all hydrocarbon groups each having from 4 to 30 carbon atoms; (4) three R^{31} 's are all aromatic hydrocarbon groups each having from 6 to 30 carbon atoms, preferably phenyl groups; (5) R^{32} is an alkyl group having at least 2 carbon atoms. Concretely, more preferred are those where R^{31} 's are all phenyl groups, and R^{32} is an isobutyl group. Most preferred is a reaction product of triphenylmethyl alcohol with triisobutylaluminium.

The reaction condition for the compound <1> and the compound <2> is not specifically defined, but is preferably as follows: They are blended in a ratio by mol, compound <1>/compound <2> falling between 1/0.1 and 1/10, more preferably between 1/0.5 and 1/2, even more preferably between 1/0.8 and 1/1.2. The reaction temperature falls between -80°C and 300°C, more preferably between -10°C and 50°C; and the reaction time falls between 0.1 minutes and 50 hours, preferably between 0.1 minutes and 3 hours. The solvent for the reaction is not also specifically defined, but is preferably the same one as that for polymerization to be effected in the presence of the catalyst.

In place of using the compound of the above-mentioned general formula as the component (C), a compound of the following (C1) and a compound of the following (C2) may be directly added to the site of catalyst production or to the

site of polymerization to form the component (C) in situ. In this case, the catalyst components are the transition metal compound (A), the compound (B) capable of reacting with a transition metal to form an ionic complex, the compounds (C1) and (C2), and optionally an alkylating agent (D).

(C1) is a compound of a general formula, $(R^{31})_3-C-OR^{33}$, or at least one selected from compounds of a general formula, $(R^{31})_3-C-OR^{33}$, $R^{34}-CO-R^{35}$ or $R^{36}-CO-OR^{37}$; and (C2) is a compound of a general formula, $Z(R^{32})_m$.

In these formulae, R^{31} represents a hydrogen atom, a halogen atom, an aliphatic hydrocarbon group having from 1 to 30 carbon atoms, an aromatic hydrocarbon group having from 6 to 30 carbon atoms, an alkoxy group having from 1 to 30 carbon atoms, an aryloxy group having from 6 to 30 carbon atoms, a thioalkoxy group having from 1 to 30 carbon atoms, a thioaryloxy group having from 6 to 30 carbon atoms, an amino group, an amido group, or a carboxyl group, R^{31} 's may be the same or different, and R^{31} 's may be optionally bonded to each other to form a cyclic structure; R^{33} , R^{34} , R^{35} , R^{36} and R^{37} each represent a hydrogen atom, a halogen atom, an aliphatic hydrocarbon group having from 1 to 30 carbon atoms, an aromatic hydrocarbon group having from 6 to 30 carbon atoms, an alkoxy group having from 1 to 30 carbon atoms, an aryloxy group having from 6 to 30 carbon atoms, a thioalkoxy group having from 1 to 30 carbon atoms, a thioaryloxy group having from 6 to 30 carbon atoms, an amino

group, an amido group, or a carboxyl group, and R^{33} , R^{34} , R^{35} , R^{36} and R^{37} may be the same or different; Z^{20} represents a metal element of Groups 2 to 13; m is an integer, indicating the valency of the metal element Z^{20} ; and R^{32} represents a hydrocarbon group.

Concretely, (C1) is at least one selected from alcohols, ethers, aldehydes, ketones, carboxylic acids and carboxylates, preferably from alcohols; and (C2) is an aluminium compound. For these, preferred are the following: (1) at least one of three R^{31} 's is an aromatic hydrocarbon group having from 6 to 30 carbon atoms; (2) three R^{31} 's are all hydrocarbon groups each having at least one carbon atom; (3) three R^{31} 's are all hydrocarbon groups each having from 4 to 30 carbon atoms; (4) three R^{31} 's for are all aromatic hydrocarbon groups each having from 6 to 30 carbon atoms, preferably phenyl groups; (5) R^{32} is an alkyl group having at least 2 carbon atoms. More concretely, the most preferred combination is (C1) of triphenylmethyl alcohol and (C2) of triisobutylaluminium.

(D) Alkylating agent:

The catalyst of the invention for production of olefinic polymers optionally contains an alkylating agent. For the details of the alkylating agent, the same as in the section of the first aspect shall directly apply also to this section.

2. Method for producing catalyst:

(1) Order of contacting constituent components:

To produce the catalyst of the invention, the order of contacting the constituent components with each other is not specifically defined. For its details, the same as in the section of the first aspect shall directly apply also to this section.

(2) Blend ratio of constituent components:

<1> Regarding the ratio by mol of the component (A) to the component (B), referred to is a case where an organoaluminium compound is used as the component (B), a compound capable of reacting with a transition metal compound to form an ionic complex. In that case, the molar ratio of the component (A) to the component (B) may fall between 1/1 and 1/10,000, but preferably between 1/10 and 1/1,000, in terms of the molar ratio to the aluminium atom in (B).

<2> Regarding the ratio by mol of the component (A) to the component (C), referred to is a case where an aluminium compound is used as the component (C). In that case, the molar ratio of the component (A) to the component (C) may fall between 1/0.5 and 1/1,000, but preferably between 1/0.8 and 1/100, more preferably between 1/1 and 1/100, in terms of the molar ratio to the aluminium atom in (C). Regarding the ratio by mol of the component (A) to the component (D), referred to is a case where an aluminium compound is used as the component (D). In that case, the molar ratio of the component (A) to the component (D) may fall between 1/0.5 and 1/1,000, but preferably between

1/1 and 1/100, in terms of the molar ratio to the aluminium atom in (D).

<3> Where the catalyst contains a combination of the component (C1) and the component (C2), but not the component (C), the molar ratio of (C1) to (C2) may fall between 1/0.1 and 1/50, precisely between 1/0.1 and 1/10, or between 1/0.5 and 1/20, or between 1/0.8 and 1/10, or between 1/0.5 and 1/2, or between 1/0.8 and 1/1.2, but most preferably between 1/0.8 and 1/10. Regarding the ratio by mol of the component (A) to the component (C2), referred to is a case where an aluminium compound is used as the component (C2). In that case, the molar ratio of the component (A) to the component (C2) may fall between 1/0.5 and 1/1,000, but preferably between 1/1 and 1/100, in terms of the molar ratio to the aluminium atom in (C2). Regarding the blend ratio of the component (D), the same as in the case <2> shall apply also to the case <3>.

(3) Condition for contacting constituent components:

To produce the catalyst, the constituent components may be contacted with each other in an inert atmosphere of nitrogen or the like, at a temperature not higher than the temperature at which the catalyst is used for polymerization. As the case may be, they may be contacted with each other at a temperature falling between -30 and 200°C.

(4) In the invention, the components constituting the catalyst, especially the component (B) may be carried on a

the same as in the section of the first aspect shall directly apply also to this section.

The amount of the catalyst to be used in the polymerization method is preferably so controlled that the component (A) may fall generally between 0.1 and 100 μ moles, but preferably between 0.5 and 25 μ moles, in one liter of the solvent used, in view of the polymerization activity and the reactor efficiency.

Regarding the polymerization condition, the pressure may fall generally between normal pressure and 2000 kg/cm²G (196 MPaG). The reaction temperature may fall generally between -50 and 250°C. For controlling the molecular weight of the polymer to be produced, the type and the amount of the catalyst components to be used and the polymerization temperature shall be suitably controlled, or hydrogen may be introduced into the polymerization system.

The invention is described in more detail with reference to the following Examples, which, however, are not intended to restrict the scope of the invention.

[Example II-1]

(1) 0.875 ml of 2 M triisobutylaluminium was added to a toluene solution of 455 mg (1.75 mmols) of triphenylmethanol at -78°C, and stirred at room temperature for 24 hours. The concentration of the component (C) thus formed was 0.1 mols/liter.

(2) 360 ml of toluene, 40 ml of 1-octene, 1.0 ml of a toluene solution of 1.0 M triisobutylaluminum, 0.1 ml of a toluene solution of N,N-dimethylanilinium tetrakis(pentafluorophenyl)borate (10 mmols/liter), and 5 ml of the component (C) prepared in the step (1) were put into a 1.6-liter autoclave equipped with a catalyst feeder tube, in that order, and heated up to 70°C. Next, ethylene was introduced into the autoclave to have a pressure of 0.785 MPaG (8 kg/cm²G). Next, 1.0 μmol of (t-butylamido)dimethyl(tetramethyl-η5-cyclopentadienyl)silane-titanium dichloride dissolved in 20 ml of toluene was put into the autoclave via the catalyst feeder tube. Ethylene was continuously introduced into the autoclave so as to have all the time the constant pressure of 8.0 kg/cm²G, and polymerized for 60 minutes. Next, methanol was added to this to stop the polymerization. A large amount of methanol was added to this, and the polymer formed was separated through filtration, and then dried at 60°C under reduced pressure for 4 hours. Thus obtained, the ethylene-1-octene copolymer weighed 46.1 g. As measured through GPC, its weight-average molecular weight was 1,270,000 in terms of ethylene, and its molecular weight distribution was 2.46. As measured through ¹H-NMR, the 1-octene content of the copolymer was 18.4 mol%.

[Comparative Example II-1]

The same process as in Example II-1 was repeated, except

that the component (C) prepared in the step of Example II-1 (1) was not added to the polymerization system in the step of Example II-1 (2). As a result, herein obtained was 35.3 g of ethylene-1-octene copolymer. As measured through GPC, its weight-average molecular weight was 1,170,000 in terms of ethylene, and its molecular weight distribution was 2.38. As measured through $^1\text{H-NMR}$, the 1-octene content of the copolymer was 16.7 mol%.

[Example II-2]

400 ml of toluene, 1.0 ml of a toluene solution of 1.0 M triisobutylaluminium, 0.1 ml of a toluene solution of tris(pentafluorophenyl)borane (10 mmols/liter), and 5 ml of the component (C) prepared in the step of Example II-1 (1) were put into a 1.6-liter autoclave equipped with a catalyst feeder tube, in that order, and heated up to 70°C. Next, ethylene was introduced into the autoclave to have a pressure of 0.785 MPaG (8 kg/cm²G). Next, 1.0 μmol of (t-butylamido)dimethyl(tetramethyl- η 5-cyclopentadienyl)silane-titaniumdimethyl dissolved in 20 ml of toluene was put into the autoclave via the catalyst feeder tube. Ethylene was continuously introduced into the autoclave so as to have all the time the constant pressure of 0.785 MPaG (8.0 kg/cm²G), and polymerized for 60 minutes. Next, methanol was added to this to stop the polymerization. A large amount of methanol was added to this, and the polymer formed was

separated through filtration, and then dried at 60°C under reduced pressure for 4 hours. Thus obtained, the ethylene polymer weighed 16.2 g. As measured through GPC, its weight-average molecular weight was 1,140,000 in terms of ethylene, and its molecular weight distribution was 1.98.

[Comparative Example II-2]

The same process as in Example II-2 was repeated, except that the component (C) prepared in the step of Example II-1 (1) was not added to the polymerization system. As a result, herein obtained was 10.5 g of ethylene polymer. As measured through GPC, its weight-average molecular weight was 1,090,000 in terms of ethylene, and its molecular weight distribution was 2.02.

[Example II-3]

The same process as in Example II-2 was repeated, except that pentamethylcyclopentadienyltitanium trimethoxide was used in place of (t-butylamido)dimethyl(tetramethyl- η 5-cyclopentadienyl)silane-titaniumdimethyl. As a result, herein obtained was 5.4 g of ethylene polymer. As measured through GPC, its weight-average molecular weight was 1,090,000 in terms of ethylene, and its molecular weight distribution was 2.02.

[Comparative Example II-3]

The same process as in Example II-3 was repeated, except that the component (C) was not added to the polymerization

system. As a result, herein obtained was 3.7 g of ethylene polymer. As measured through GPC, its weight-average molecular weight was 1,090,000 in terms of ethylene, and its molecular weight distribution was 2.02.

[Example II-4]

(1) Preparation of component (C):

0.875 ml of 2 M triisobutylaluminium was added to a toluene solution of 1.75 mmols of tricyclohexylmethanol (from Across) at -78°C, and stirred at room temperature for 24 hours. The concentration of the component (C) thus formed was 0.1 mols/liter.

Through ¹H-NMR, the product was confirmed to have the component (C) formed therein.

(2) Polymerization of ethylene:

Ethylene was polymerized in the same manner as in Example II-1, except that the compound prepared in the above step (1) was used as the component (C) in place of that prepared in the step of Example II-1 (1). As a result, herein obtained was 8.7 g of ethylene polymer. As measured through GPC, its weight-average molecular weight was 1,100,000 in terms of ethylene, and its molecular weight distribution was 2.05.

[Example II-5]

The same process as in Example II-2 was repeated, except that dimethylsilylenebisbenzoindenylzirconium dichloride was used in place of (t-butylamido)dimethyl(tetramethyl-η5-

cyclopentadienyl)silane-titaniumdimethyl, and that propylene was used in place of ethylene. As a result, herein obtained was 34.1 g of propylene polymer. As measured through GPC, its weight-average molecular weight was 1,050,000, and its molecular weight distribution was 2.03.

[Comparative Example II-4]

The same process as in Example II-5 was repeated, except that the component (C) was not added to the polymerization system. As a result, herein obtained was 16.4 g of propylene polymer. As measured through GPC, its weight-average molecular weight was 980,000, and its molecular weight distribution was 2.04.

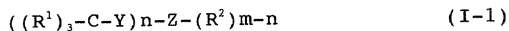
INDUSTRIAL APPLICABILITY

The catalysts of the invention for polymerization of olefins have the advantages of enhancing the polymerization activity and reducing the amount of the promoter such as oxygen-containing compounds, etc. Using them, therefore, olefinic polymers can be produced efficiently and inexpensively.

CLAIMS

1. A catalyst for polymerization of olefins, which comprises:

- (A) a transition metal compound,
- (B) an oxygen-containing compound,
- (C) a compound of a general formula (I-1):



wherein R^1 represents an aromatic hydrocarbon group having from 6 to 30 carbon atoms, an alkoxy group having from 1 to 30 carbon atoms, an aryloxy group having from 6 to 30 carbon atoms, a thioalkoxy group having from 1 to 30 carbon atoms, a thioaryloxy group having from 6 to 30 carbon atoms, an amino group, an amido group, or a carboxyl group, R^1 's may be the same or different, and R^1 's may be optionally bonded to each other to form a cyclic structure; Y represents an element of Group 16; Z represents a metal element of Groups 2 to 13; R^2 represents a hydrocarbon group; m is an integer, indicating the valency of the metal element Z; and n is an integer of from 1 to (m-1),

and optionally,

- (D) an alkylating agent.

2. The catalyst as claimed in claim 1 for polymerization of olefins, wherein, in (C), Y is oxygen and Z is aluminium.

3. The catalyst as claimed in claim 1 for

polymerization of olefins, wherein the compound (C) is a reaction product of a compound of a general formula, $(R^1)_3C-OR^3$, and a compound of a general formula, $Z(R^2)_m$:

in which R^1 represents an aromatic hydrocarbon group having from 6 to 30 carbon atoms, an alkoxy group having from 1 to 30 carbon atoms, an aryloxy group having from 6 to 30 carbon atoms, a thioalkoxy group having from 1 to 30 carbon atoms, a thioaryloxy group having from 6 to 30 carbon atoms, an amino group, an amido group, or a carboxyl group, R^1 's may be the same or different, and R^1 's may be optionally bonded to each other to form a cyclic structure; R^3 represents a hydrogen atom, a halogen atom, an aliphatic hydrocarbon group having from 1 to 30 carbon atoms, an aromatic hydrocarbon group having from 6 to 30 carbon atoms, an alkoxy group having from 1 to 30 carbon atoms, an aryloxy group having from 6 to 30 carbon atoms, a thioalkoxy group having from 1 to 30 carbon atoms, a thioaryloxy group having from 6 to 30 carbon atoms, an amino group, an amido group, or a carboxyl group; Z represents a metal element of Groups 2 to 13; m is an integer, indicating the valency of the metal element Z ; and R^2 represents a hydrocarbon group.

4. A catalyst for polymerization of olefins, which comprises:

- (A) a transition metal compound,
- (B) an oxygen-containing compound,

R¹'s is an aromatic hydrocarbon group having from 6 to 30 carbon atoms.

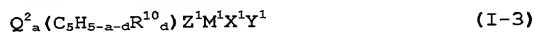
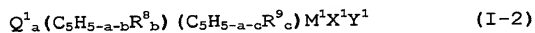
6. The catalyst for polymerization of olefins as claimed in any of claims 1 to 4, wherein three R¹'s are all aromatic hydrocarbon groups each having from 6 to 30 carbon atoms.

7. The catalyst for polymerization of olefins as claimed in any of claims 1 to 4, wherein three R¹'s are all phenyl groups.

8. The catalyst for polymerization of olefins as claimed in any of claims 1 to 7, wherein R² is an alkyl group having at least 2 carbon atoms.

9. The catalyst for polymerization of olefins as claimed in any of claims 3 to 8, wherein Z is aluminium.

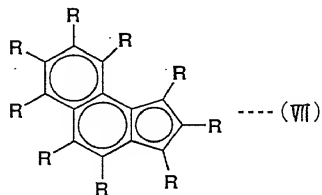
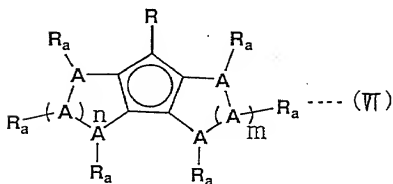
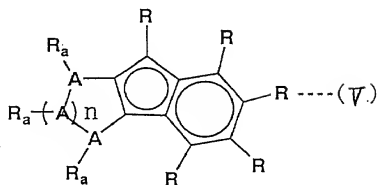
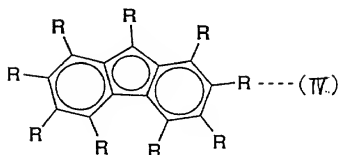
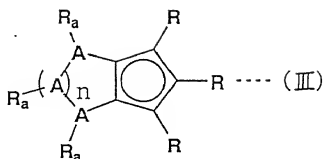
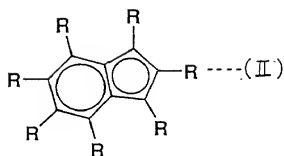
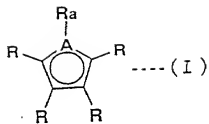
10. The catalyst for polymerization of olefins as claimed in any of claims 1 to 9, wherein the transition metal compound (A) is represented by any of the following general formulae (I-2) to (I-6):



in which Q¹ represents a bonding group that crosslinks the two conjugated five-membered cyclic ligands (C₅H_{5-a-b}R⁸_b) and

$(C_5H_{5-a-c}R^9_c)$; Q^2 represents a bonding group that crosslinks the conjugated five-membered cyclic ligand $(C_5H_{5-a-d}R^{10}_d)$ and the group Z^1 ; R^8 , R^9 , R^{10} and R^{11} each represent a hydrocarbon group, a halogen atom, an alkoxy group, a silicon-containing hydrocarbon group, a phosphorus-containing hydrocarbon group, a nitrogen-containing hydrocarbon group, or a boron-containing hydrocarbon group; and a plurality of these groups, if any, may be the same or different, and may be bonded to each other to form a cyclic structure; a represents 0, 1 or 2; b, c and d each represent an integer of from 0 to 5 when a = 0, or an integer of from 0 to 4 when a = 1, or an integer of from 0 to 3 when a = 2; e is an integer of from 0 to 5; M^1 represents a transition metal of Groups 4 to 6 of the Periodic Table; M^2 represents a transition metal of Groups 8 to 10 of the Periodic Table; L^1 and L^2 each represent a coordination-bonding ligand; X^1 , Y^1 , Z^1 , W^1 and U^1 each represent a covalent-bonding or ionic-bonding ligand; and L^1 , L^2 , X^1 , Y^1 , Z^1 , W^1 and U^1 may be bonded to each other to form a cyclic structure.

11. The catalyst for polymerization of olefins as claimed in claim 10, wherein, in the transition metal compound (A) of formula (I-4), the group $(C_5H_{5-e}R^{11}_e)$ is represented by any of the following general formulae (I) to (VII):



wherein A represents an element of Group 13, 14, 15 or 16, and plural A's may be the same or different; R represents a hydrogen atom, a halogen atom, an aliphatic hydrocarbon group having from 1 to 30 carbon atoms, an aromatic hydrocarbon group having from 6 to 30 carbon atoms, an alkoxy group having from 1 to 30 carbon atoms, an aryloxy group having from 6 to 30 carbon atoms, a thioalkoxy group having from 1 to 30 carbon atoms, a thioaryloxy group having from

6 to 30 carbon atoms, an amino group, an amido group, a carboxyl group, or an alkylsilyl or alkylsilylalkyl group having from 3 to 30 carbon atoms, and R's may be the same or different, and may be optionally bonded to each other to form a cyclic structure; a represents 0, 1 or 2; and n and m each represent an integer of at least 1.

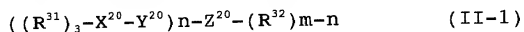
12. A method for producing olefinic polymers, which comprises polymerizing olefins in the presence of the polymerization catalyst of any of claims 1 to 11.

13. A catalyst for polymerization of olefins, which comprises:

(A) a transition metal compound,

(B) a compound capable of reacting with a transition metal compound to form an ionic complex,

(C) a compound of a general formula (II-1):



wherein R^{31} represents a hydrogen atom, a halogen atom, an aliphatic hydrocarbon group having from 1 to 30 carbon atoms, an aromatic hydrocarbon group having from 6 to 30 carbon atoms, an alkoxy group having from 1 to 30 carbon atoms, an aryloxy group having from 6 to 30 carbon atoms, a thioalkoxy group having from 1 to 30 carbon atoms, a thioaryloxy group having from 6 to 30 carbon atoms, an amino group, an amido group, or a carboxyl group, R^{31} 's may be the same or different, and R^{31} 's may be optionally bonded

to each other to form a cyclic structure; X^{20} represents an element of Group 14; Y^{20} represents an element of Group 16; Z^{20} represents a metal element of Groups 2 to 13; R^{32} represents a hydrocarbon group; m is an integer, indicating the valency of the metal element Z^{20} ; and n is an integer of from 1 to $(m-1)$,

and optionally,

(D) an alkylating agent.

14. The catalyst for polymerization of olefins as claimed in claim 13, wherein, in (C), Y^{20} is oxygen and Z^{20} is aluminium.

15. The catalyst for polymerization of olefins as claimed in claim 13, wherein the compound (C) is a reaction product of a compound of a general formula, $(R^{31})_3-C-OR^{33}$, and a compound of a general formula, $Z^{20}(R^{32})_m$:

in which R^{31} represents a hydrogen atom, a halogen atom, an aliphatic hydrocarbon group having from 1 to 30 carbon atoms, an aromatic hydrocarbon group having from 6 to 30 carbon atoms, an alkoxy group having from 1 to 30 carbon atoms, an aryloxy group having from 6 to 30 carbon atoms, a thioalkoxy group having from 1 to 30 carbon atoms, a thioaryloxy group having from 6 to 30 carbon atoms, an amino group, an amido group, or a carboxyl group, R^{31} 's may be the same or different, and R^{31} 's may be optionally bonded to each other to form a cyclic structure; Z^{20} represents

group, an amido group, or a carboxyl group, R^{31} 's may be the same or different, and R^{31} 's may be optionally bonded to each other to form a cyclic structure; R^{33} represents a hydrogen atom, a halogen atom, an aliphatic hydrocarbon group having from 1 to 30 carbon atoms, an aromatic hydrocarbon group having from 6 to 30 carbon atoms, an alkoxy group having from 1 to 30 carbon atoms, an aryloxy group having from 6 to 30 carbon atoms, a thioalkoxy group having from 1 to 30 carbon atoms, a thioaryloxy group having from 6 to 30 carbon atoms, an amino group, an amido group, or a carboxyl group, and may be the same or different,

(C2) a compound of a general formula, $Z^{20}(R^{32})_m$, wherein Z^{20} represents a metal element of Groups 2 to 13; m is an integer, indicating the valency of the metal element Z^{20} ; and R^{32} represents a hydrocarbon group,

and optionally,

(D) an alkylating agent.

17. The catalyst for polymerization of olefins as claimed in any of claims 13 to 16, wherein at least one of three R^{31} 's is an aromatic hydrocarbon group having from 6 to 30 carbon atoms.

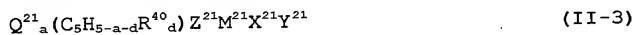
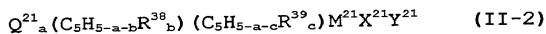
18. The catalyst for polymerization of olefins as claimed in any of claims 13 to 16, wherein three R^{31} 's are all aromatic hydrocarbon groups each having from 6 to 30 carbon atoms.

19. The catalyst for polymerization of olefins as claimed in any of claims 13 to 16, wherein three R³¹'s are all phenyl groups.

20. The catalyst for polymerization of olefins as claimed in any of claims 13 to 19, wherein R³² is an alkyl group having at least 2 carbon atoms.

21. The catalyst for polymerization of olefins as claimed in any of claims 15 to 20, wherein Z is aluminium.

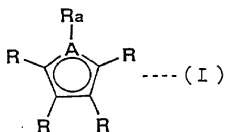
22. The catalyst for polymerization of olefins as claimed in any of claims 13 to 21, wherein the transition metal compound (A) is represented by any of the following general formulae (II-2) to (II-6):

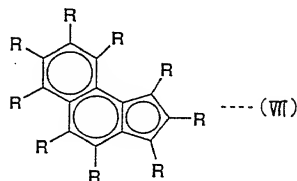
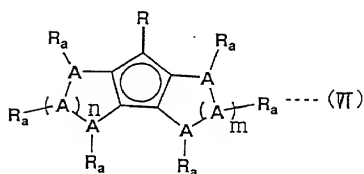
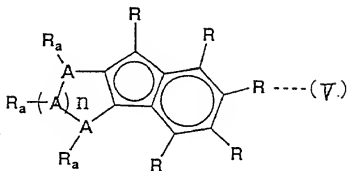
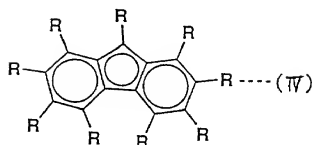
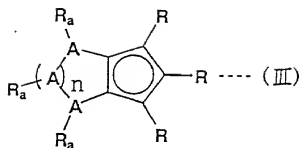
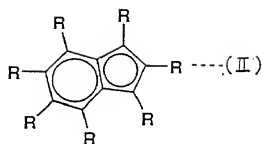


in which Q²¹ represents a bonding group that crosslinks the two conjugated five-membered cyclic ligands (C₅H_{5-a-b}R³⁸_b) and (C₅H_{5-a-c}R³⁹_c); Q²² represents a bonding group that crosslinks the conjugated five-membered cyclic ligand (C₅H_{5-a-d}R⁴⁰_d) and the group Z²¹; R³⁸, R³⁹, R⁴⁰ and R⁴¹ each represent a hydrocarbon group, a halogen atom, an alkoxy group, a silicon-containing hydrocarbon group, a phosphorus-containing hydrocarbon group, a nitrogen-

containing hydrocarbon group, or a boron-containing hydrocarbon group; and a plurality of these groups, if any, may be the same or different, and may be bonded to each other to form a cyclic structure; a represents 0, 1 or 2; b, c and d each represent an integer of from 0 to 5 when a = 0, or an integer of from 0 to 4 when a = 1, or an integer of from 0 to 3 when a = 2; e is an integer of from 0 to 5; M^{21} represents a transition metal of Groups 4 to 6 of the Periodic Table; M^{22} represents a transition metal of Groups 8 to 10 of the Periodic Table; L^{21} and L^{22} each represent a coordination-bonding ligand; X^{21} , Y^{21} , Z^{21} , W^{21} and U^{21} each represent a covalent-bonding or ionic-bonding ligand; and L^{21} , L^{22} , X^{21} , Y^{21} , Z^{21} , W^{21} and U^{21} may be bonded to each other to form a cyclic structure.

23. The catalyst for polymerization of olefins as claimed in claim 22, wherein, in the transition metal compound (A) of formula (II-4), the group $(C_5H_5-\sigma-R^{41})$ is represented by any of the following general formulae (I) to (VII):





wherein A represents an element of Group 13, 14, 15 or 16, and plural A's may be the same or different; R represents a hydrogen atom, a halogen atom, an aliphatic hydrocarbon group having from 1 to 30 carbon atoms, an aromatic hydrocarbon group having from 6 to 30 carbon atoms, an alkoxy group having from 1 to 30 carbon atoms, an aryloxy group having from 6 to 30 carbon atoms, a thioalkoxy group having from 1 to 30 carbon atoms, a thioaryloxy group having from 6 to 30 carbon atoms, an amino group, an amido group, a carboxyl group, or an alkylsilyl or alkylsilylalkyl group having from 3 to 30 carbon atoms, and R's may be the same or different, and may be optionally bonded to each other

ABSTRACT

The invention includes catalysts for polymerization of olefins, comprising (A) a transition metal compound, (B) an oxygen-containing compound, or a compound capable of reacting with a transition metal compound to form an ionic complex, (C) a specific compound such as a reaction product of triphenylmethyl alcohol and triisobutylaluminum, and optionally (D) an alkylating agent, and methods for producing olefinic polymers in the presence of such catalysts. The catalysts have the advantages of enhancing the polymerization activity and reducing the amount of the promoter such as oxygen-containing compounds, etc. Using them, olefinic polymers can be produced efficiently and inexpensively.

Declaration, Power Of Attorney and Petition

Page 1 of 3

We (I) the undersigned inventor(s), hereby declare(s) that:

My residence, post office address and citizenship are as stated below next to my name,

We (I) believe that we are (I am) the original, first, and joint (sole) inventor(s) of the subject matter which is claimed and for which a patent is sought on the invention entitled

CATALYST FOR OLEFIN POLYMERIZATION AND PROCESS FOR PRODUCING OLEFIN
POLYMER

the specification of which

- ☐ is attached hereto.
- ☐ was filed on _____ as
Application Serial No. _____
and amended on _____
- ☒ was filed as PCT international application
Number PCT/JP00/00229
on January 19, 2000,
and was amended under PCT Article 19
on _____ (if applicable).

We (I) hereby state that we (I) have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above.

We (I) acknowledge the duty to disclose information known to be material to the patentability of this application as defined in Section 1.56 of Title 37 Code of Federal Regulations.

We (I) hereby claim foreign priority benefits under 35 U.S.C. § 119(a)-(d) or § 365(b) of any foreign application(s) for patent or inventor's certificate, or § 365(a) of any PCT International application which designated at least one country other than the United States, listed below and have also identified below, by checking the box, any foreign application for patent or inventor's certificate, or PCT International application having a filing date before that of the application on which priority is claimed. Prior Foreign Application(s)

Application No	Country	Day/Month/Year	Priority Claimed
<u>11-022075</u>	<u>Japan</u>	<u>29/ 1/1999</u>	<input checked="" type="checkbox"/> Yes <input type="checkbox"/> No
<u>11-146305</u>	<u>Japan</u>	<u>26/ 5/1999</u>	<input checked="" type="checkbox"/> Yes <input type="checkbox"/> No
_____	_____	_____	<input type="checkbox"/> Yes <input type="checkbox"/> No
_____	_____	_____	<input type="checkbox"/> Yes <input type="checkbox"/> No

We (I) hereby claim the benefit under Title 35, United States Code, § 119(e) of any United States provisional application(s) listed below.

(Application Number)

(Filing Date)

(Application Number)

(Filing Date)

We (I) hereby claim the benefit under 35 U.S.C. § 120 of any United States application(s), or § 365 (c) of any PCT International application designating the United States, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States or PCT International application in the manner provided by the first paragraph of 35 U.S.C. § 112, I acknowledge the duty to disclose information which is material to patentability as defined in 37 CFR § 1.56 which became available between the filing date of the prior application and the national or PCT International filing date of this application.

Application Serial No.

Filing Date

Status (pending, patented,
abandoned)

And we (I) hereby appoint: Norman F. Oblon, Reg.No.24,618; Marvin J. Spivak, Reg.No.24,913; C. Irvin McClelland, Reg.No.21,124; Gregory J. Maier, Reg.No.25,599; Arthur I. Neustadt, Reg.No.24,854; Richard D. Kelly, Reg.No.27,757; James D. Hamilton, Reg.No.28,421; Eckhard H. Kuesters, Reg.No.28,870; Robert T. Pous, Reg.No.29,099; Charles L. Gholz, Reg.No.26,395; Vincent J. Sunderdick, Reg.No.29,004; William E. Beaumont, Reg.No.30,996; Robert F. Gnuse, Reg.No.27,295; Jean-Paul Lavalleye, Reg.No.31,451; Stephen G. Baxter, Reg.No.32,884; Robert W. Hahl, Reg.No.33,893; Richard L. Chinn, Reg.No.36,379; Steven P. Weihrouch, Reg.No.32,829; John T. Goolkasian, Reg.No.26,142; Richard L. Chinn, Reg.No.34,305; Steven E. Lipman, Reg.No.30,001; Carl E. Schlier, Reg.No.34,426; James J. Kulbaski, Reg.No.34,648; Richard A. Neifeld, Reg.No.35,299; J.Derek Mason, Reg. No.35,270; Surinder Sachar, Reg. No.34,423; Christina M. Gadiano, Reg. No.37,628; Jeffrey B. McIntyre, Reg.No.36,867; Paul E. Rauch, Reg.No.38,591; William T. Enos, Reg.No.33,128; and Micheal E. McCabe, Jr., Reg.No.37,182; our(my) attorneys, with full powers of substitution and revocation, to prosecute this application and to transact all business in the Patent Office connected therewith; and we(I) hereby request that all correspondence regarding this application be sent to the firm of OBLON, SPIVAK, MCCLELLAND, MAIER & NEUSTADT, P.C., whose Post Office Address is: Fourth Floor, 1755 Jefferson Davis Highway, Arlington, Virginia 22202.

We (I) declare that all statements made herein of our (my) own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

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